Attosecond Physics – the first decade

On the route to tiny time scales

Deep inside matter, our ideas of time lose their validity. Light flashes which last only a few millionths to billionths of a billionth second snatch from the microcosm its closely guarded secrets: Electron movements become visible. Quantum phenomena can be examined in real time. The control of elementary particles comes within reach. Responsible for all that is the young science of attosecond physics, "born" in 2001, when Prof. Ferenc Krausz succeeded in producing and measuring light flashes which last less than one femtosecond. Krausz founded the Laboratory for Attosecond Physics (LAP) which is located at the Max Planck Institute for Quantum Optics in Garching, Germany. Here is the history of how Attosecond Physics has been developed in LAP over the last ten years.

Ever smaller, ever faster

When scientists started to "take pictures" of the movements of atoms in molecules during chemical reactions in the beginning 1990s, this had little to do with classical photography. The principle, however, remained the same: A short exposure time is used to produce sharp pictures. Structures of atoms and molecules change within femtoseconds, i.e., on a scale one thousand times longer than an attosecond. This is why an appropriate exposure time in the range of femtoseconds became necessary. Scientists found it in laser technology thanks to femtosecond laser flashes.

In spite of groundbreaking success and the Nobel Prize for Egyptian scientist Ahmed Zewail (in 1999), some questions remained unanswered. What happens inside the electron shell which surrounds the atomic nucleus? How can movements of individual electrons be tracked? Are there ways to verify our concepts of atomic structures? In this context, it appeared obvious that processes in subatomic dimensions proceed faster than in comparably slow-moving atoms and molecules.

Electrons propagate within attoseconds. For "taking pictures" of such objects, one must be as quick. Attosecond physics makes this possible. Thus, pursuing far shorter exposure times, even smaller fractions of seconds, and spectacular images from microcosm is getting under way. In the late 1990s Prof. Paul Corkum and his group proposed for the first time a method for producing and measuring attosecond pulses of light after studying the interaction of intense laser radiation with atoms and molecules.

2001: A new dimension of time

New dimensions of time intervals are now at reach. At Vienna University of Technology, Ferenc Krausz and his team – for the first time – succeed in producing light flashes which last only attoseconds. For this, the scientists focus laser pulses lasting seven femtoseconds on neon inert gas atoms. Their electrons absorb the light's energy and subsequently emit it as X-ray flashes. The scientists then isolate such flashes which last just 650 attoseconds. Attosecond physics was "born" (Nature, November 29, 2001).

2002: The world's highest speed camera

For more than a decade it has already been possible to explore the movements of atoms and molecules during chemical reactions by means of femtosecond laser pulses. However, the considerably smaller electrons are much quicker. In a hydrogen atom for example, an electron orbits the nucleus within 24 attoseconds.

By means of attosecond flashes it is now possible to observe such movements of electrons inside an atom. X-ray flashes are sufficiently energetic to propagate deep into krypton atoms. Scientists use such flashes to knock out an electron from the innermost shell. Thus, the flash ionizes the atom. Ionization is an ordinary phenomenon in chemistry but electrons are usually removed from the outer shells.

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However, the attosecond flash extracts an electron from the atom's innermost shell. In a way, it sends the atom into a state of shock. Remaining electrons try to fill up the hole as quickly as possible. During this chaotic process, the krypton atom releases a second electron which is referred to as Auger electron. The Auger electron remains outside the atom until the hole inside the atom is refilled.

This gives physicists an important piece of information. The Auger electron reveals how long it takes to fill up the hole inside the atom with a new electron (Nature, December 19, 2002), (Figure 1).

2003: Perfect control over light waves

Light as a medium is difficult to handle. It propagates in all directions, features numerous wavelengths and therefore a conglomerate of electromagnetic fields which is hard to grasp. For centuries scientists have been trying to find ways to better control light. A breakthrough was made by Theodor Hänsch and his team in the 1990s. They developed a frequency comb. The tool allows for precise measurement of the frequency of light via laser. The new technology opens the door to control the course of the electric field of laser pulses.

The physicists use such laser pulses to produce soft X-ray attosecond light pulses (approx. 50 nanometers wavelength), lasting 500 attoseconds. For the first time, the attosecond light flashes can be produced identically and timed precisely via the laser pulses. Improvements in attosecond technology allow for in-depth analyses of the behavior of tiny particles in microcosm such as rearrangements of electrons in molecules (Nature, February 6, 2003).

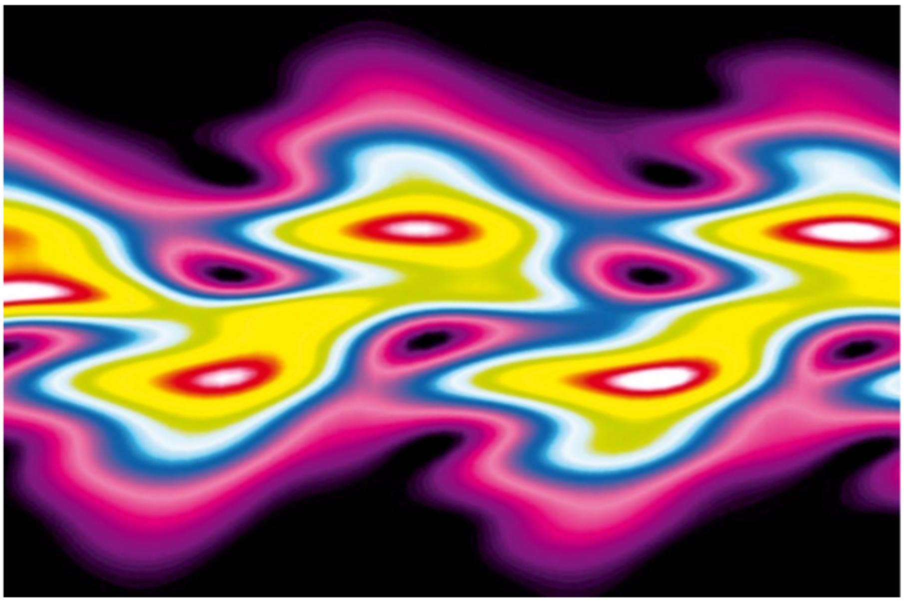

FIG. 1: **This image shows how electron movements are distributed in the atom after an attosecond light flash has extracted an electron from the atom's innermost shell.** (Source: LAP)

2004: Time captured in a picture

How can attoseconds be measured? The question remains unanswered for many years. Finally, classic mechanics provides a solution via a so-called streak camera. In the 19th century, physicists used this technique to measure the duration of light flashes produced in electric discharges. The principle was delivered by physicist Charles Wheatstone (1802–1875). He invented the first streak camera. Wheatstone used a rotating mirror. The mirror reflected the impinging light from a discharge to different spatial positions. The length of the picture and the angular velocity of the mirror were used to calculate the duration of the flash. Wheatstone discovered that some flashes lasted less than a millionth of a second.

In attosecond physics, time intervals are displayed on an energy scale rather than on a screen. The technique is referred to as "light-field-controlled streak camera". The rotating mirrors of the 19th century are nowadays replaced by the electrical field of light waves.

In this technique, an attosecond flash is accompanied by a second laser pulse. The flash knocks out electrons from a sample. The electrons are captured by the second femtosecond laser pulse. This laser pulse replaces the rotating mirrors used in Charles Wheatstone's experiments. The laser light is made up of a few oscillations of its electric field. Depending on when the escaping electrons are captured by the corresponding electric field, they accelerate differently. A detector records this behavior and thus measures the duration of the attosecond flash.

Two prerequisites apply for designing a light streak camera. The oscillations of the deflecting laser light field in the visible spectrum (300 to 780 nanometers wavelength) need to be controlled precisely. For this, the timed X-ray attosecond flash has to be exactly synchronized to the visible light (Figure 2).

2007: On the 100-attosecond barrier

Durations of light flashes are cut continuously. By then, they had reached 100 attoseconds. This development takes place due to improvements in laser technologies. Physicists are now able to produce laser pulses made up of only few wave oscillations. Usually, light requires several femtoseconds for one wave cycle. Light changes its direction approximately 100 trillion times per second. In femtosecond pulses, this happens only a few times.

Furthermore, scientists now produce pulses with extremely high energy levels. More than half of the power is released within a single wave cycle. This lasts about three femtoseconds. Massive forces are exerted in such a process. Noble gas atoms exposed to the extreme light pulses emit a new generation of X-ray attosecond flashes. Such flashes contain more than a million photons. They are short enough to capture the movements of electrons orbiting in molecular orbitals. Thus, real-time observation

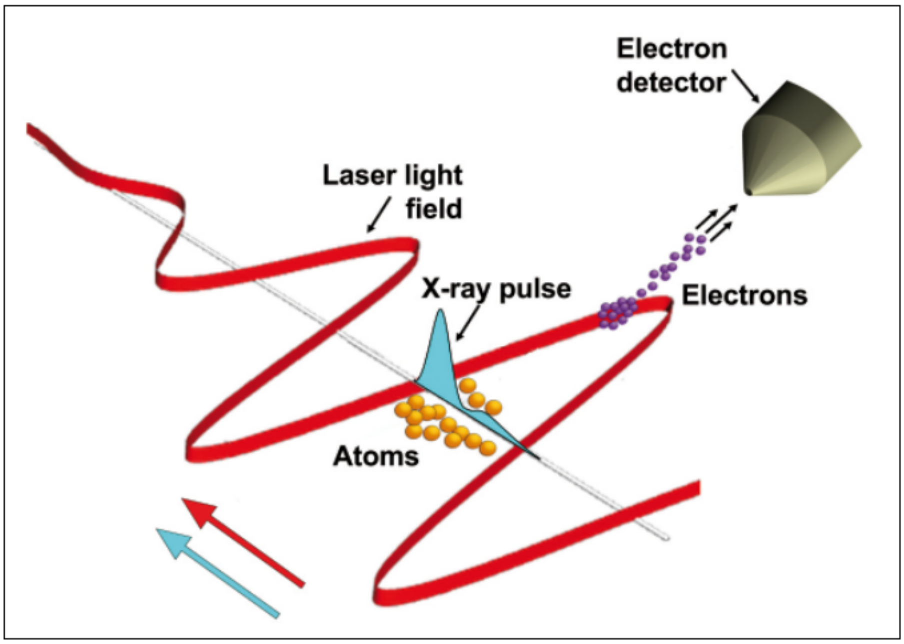

FIG. 2: **Light-field-controlled streak camera. The principle of operation is described in the text.** (Source: LAP)

FIG. 3: **Nature featured the first measurement of electron movements inside a solid on the cover of the issue of 25th October 2008.**

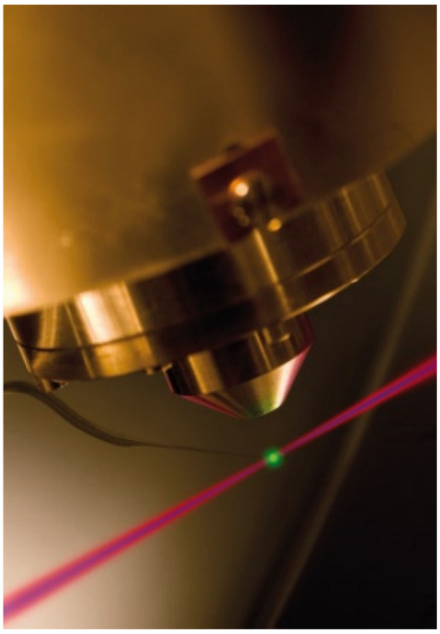

FIG. 4: **A combination of UV pulses and an IR beam allows observation of the motion of electrons in atoms.** (Source: LAP)

THE INSTITUTE

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Garching, Germany

The mission of the Attosecond Physics Laboratory is to advance attosecond science and promote its proliferation. We develop broadband light sources emitting waves with controlled oscillations of electric and magnetic fields. They provide the force for steering low-energy electrons in atomic systems as well as high-energy electrons travelling at the speed of light.

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of electrons is possible. New insights, e.g. as to how formation or disintegration of molecules can be explained, are at reach (Science, August 10, 2007).

2007: Racing to the surface

How fast do electrons propagate through a crystal? Adrian Cavalieri and his colleagues attempted to solve this puzzle. For this, they sent a light pulse of 300 attoseconds onto a tungsten crystal. Simultaneously, the same sample is exposed to a focused, infrared femtosecond pulse made up of less than two oscillations.

The attosecond pulse penetrates the crystal. Here, it releases two kinds of electrons at the same time: conduction band and core electrons. Conduction band electrons are loosely bound charge carriers which are responsible for electric conductivity. Core electrons are bound solidly to the atomic core. Both types of electrons race through several atomic layers of the crystal simultaneously but at different velocities. Conduction band electrons travel faster than core electrons. When reaching the surface, both types of particles are captured by the infrared laser pulse and modified such that they become detectable by a particle detector. The electric field of the pulse serves as a kind of stopwatch for the race. The scientists discover that the conduction band electrons reach the surface of the crystal 110 attoseconds before the core electrons do. For the first time, Adrian Cavalieri and his colleagues measure electron movements inside a solid (Nature, October 25, 2007), (Figure 3).

2008: Breaking the 100-attosecond barrier

For the first time, scientists succeed in creating light flashes which last less than 100 attoseconds. This is possible due to the developments in the technologies for producing such flashes. As previously, physicists use the electric field of laser pulses in the near infrared spectrum. During the laser pulse, this field performs hardly more than a single intense oscillation with a period of approx. 2.5 femtoseconds. That means that the light wave contains only two high wave crests with a deep valley in between. The power that the electric light field exerts on the electrons is strongest at the top of the ridges and at the deepest spot of the valley. The electric light field removes electrons from the noble gas atoms turning them into ions. Due to the oscillation of the light field, the force changes its direction and hurls the electrons back to the ions after a short period of time. When the free electrons impinge they evoke attosecond oscillations which in turn cause light flashes in the range of attoseconds. The flashes are pulses of extreme ultraviolet light (XUV, approx. 10 to 20 nanometers wavelength).

For the first time, the controlled production of a single powerful light oscillation allows for releasing electrons precisely three times within an individual laser pulse. When returning to the ion core, each particle emits a laser flash. Thus, one laser pulse produces three attosecond flashes. One of

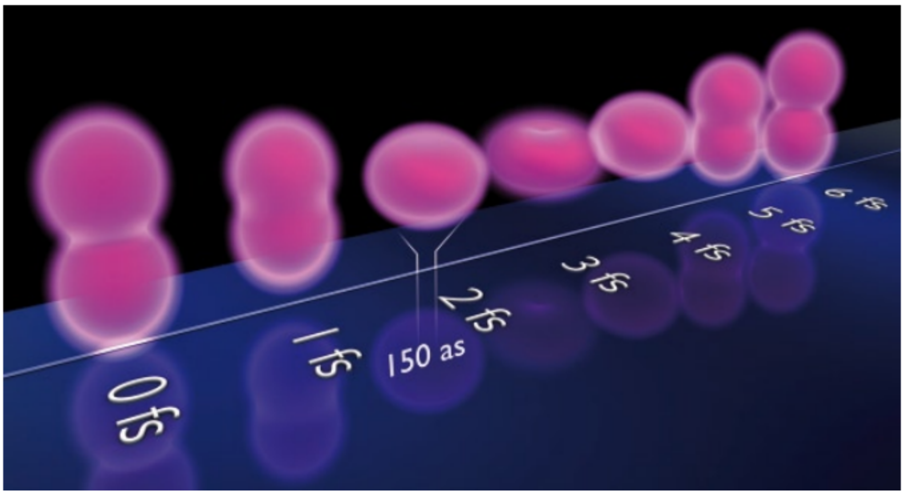

FIG. 5: **Snapshots of the oscillatory motion of a valence electron inside an atomic ion, reconstructed from attosecond pump–probe measurements.** (Source: Christian Hackenberger/LAP)

the three flashes is particularly intense. It contains more than 100 million photons within 80 attoseconds. The scientists filter out this pulse with special X-ray mirrors developed by Ulf Kleineberg's team (Science, June 20, 2008).

2010: In the footsteps of Albert Einstein

As attosecond experiments reveal, electrons in atoms do not react spontaneously when they are struck by light. This allows physics to gain new insights into the phenomenon of photoemission, discovered by Albert Einstein more than 100 years ago. To date, excitation and photoemission of electrons in atoms due to light are still among the most prominent phenomena in quantum physics. Photoemission means that electrons in atoms are excited by light; at sufficiently high energy levels, the particles leave their atom. Initially scientists thought that the electron movement starts immediately after the atom is hit by the light beam.

This assumption is under investigation by Martin Schultze and his team of attosecond physicists. For this, they focus light pulses onto neon atoms. As the experiments show, electrons on different positions (orbitals) of the atomic shell that are hit by the light pulse simultaneously, leave the atom with delays of several ten attoseconds.

In their experiments, the physicists focus strong laser pulses of about four femtoseconds onto noble gas atoms. For this, the scientists synchronize an additional light flash of just under 180 attoseconds. The attosecond flashes are used to release the electrons from their orbitals. The flashes cause either the outer 2p or inner 2s orbital (closer to the nucleus) to release electrons. The synchronized laser pulses are used to record as to when this occurs.

The physicists found out that in spite of simultaneous excitation, the electrons leave the noble gas atom with an offset of 20 attoseconds. Thus, one of the electrons is ahead of the other.

Theory is needed to explain this offset. Theorists around Vladislav Yakovlev verified the effect. However, they predict a timely offset of five attoseconds. The discrepancy is probably due to the complex nature of a neon atom. Apart from the nucleus, it consists of ten electrons. Unfortunately, computing time for the complete atomic model exceeds the capacity of supercomputers. At least calculations reveal the most probable cause for the offset of the electrons. The scientists think that the electrons not only interact with their atomic nucleus but also influence each other. Contrary to the previous assumptions, electron-electron interaction seems to be the reason why an electron hit by the incoming light wave is released by its fellow electrons to leave the atom only after a delay (Figure 4).

FIG. 6: **A light field synthesizer divides incident coherent white light into three colors and modifies it. With this, "white" laser pulses were generated for the first time. Their field can be sculpted on time scales shorter than an optical cycle.** (Source: T. Naeser/LAP)

2010: Pulsating emptiness

In the quantum world, holes pulsate. As attosecond physics proves, absolute emptiness indeed develops a life of its own. In their experiments, a team around Eleftherios Goulielmakis focuses laser pulses from the visible spectrum onto krypton atoms. The light pulses, lasting less than four femtoseconds, each knock out an electron from the outer electron shell of an atom. The missing electron, detached by the laser pulse, causes the atom to convert to an ion with positive charge. At the location where the electron escapes from the atom, it leaves a positively charged hole. In the view of quantum mechanics, this free spot in the atom continues to pulsate as a so-called quantum beat.

This pulsing is recorded, i.e. photographed, with a second pulse of extreme ultraviolet light lasting about 150 attoseconds. As it turns out, the position of the hole in the ion (i.e. of the positively charged location) moves back and forth periodically between an elongated club-like and a compact contracted shape with a period of approximately six femtoseconds. Thus, for the first time, the scientists succeeded in observing directly the change in the charge distribution inside an atom (Nature, August 5, 2010), (Figure 5). In the same year a group around Prof. Anne L'Huillier from Lund University and Prof. Marc Vrakking director of the Max Born Institute for Nonlinear Optics and Short Pulse Spectroscopy in Berlin succeeded in measuring for the first time electron movements in molecules.

2011: Custom light waves

For controlling electrons waveforms of light need to be customized on a scale within its oscillation period. This requires ultra-white laser light which contains additionally, besides all colors of visible light, infrared and ultraviolet waves. Also, a device is needed for combining such waves oscillating at different frequencies. In 2011, LAP scientists developed what they call a light wave synthesizer which allows for controlling the oscillation behavior of light with unprecedented precision. Using this technology, the scientists were able to create completely new waveforms within the pulses. Now, such pulses can consist of less than one complete oscillation while lasting only about two femtoseconds. To date, these are the shortest pulses ever produced with visible light (Figure 6).

Timely characteristics of light power are now controllable on the attosecond scale. Thus, the technology aims at precisely controlling electron movements for the first time. Since all of the energy of the electromagnetic field is concentrated into a tiny temporal window, the new tool allows for exciting processes within a femtosecond and opens the path for attosecond excitation-detection experiments (Science, October 14, 2011).