Professor Dr. Ulf Kleineberg describes his research into effecting changes to the characteristics of nanoscale materials through light pulsed at attosecond intervals.

What prompted your interest in nanophotonics?

My interest in nanophotonics was triggered by the combination of two fascinating areas of physics: understanding the nanoworld with its unique properties of matter which may differ from those in the macroscopic world, and the utilisation of photons to investigate and eventually control these properties.

Could you outline some of the attosecond tools your laboratory has developed?

The ultrafast aspect of nanophotonics is twofold: our scientific ‘tools’ are light waves whose fastest transients are given by the oscillating electromagnetic field. Modern ultrafast laser systems, such as the one we operate in our lab, can deliver very short light pulses which only last about one oscillation period of that field. We are using this rapid increase (or decrease) of the light field to excite nanostructures and steer electronic processes within a time period of less than one femtosecond, which is the attosecond time regime.

The second important aspect of nanophotonics stems from the fact that the electrons in nanosystems respond very quickly to a laser light field. As an example, free electrons in a metal can be collectively driven and accelerated back and forth by a light wave, giving rise to collective phenomena like the launching of a plasmonic wave.

In order to ‘see’ these fast-moving electrons on an attosecond time scale, we have to develop tools which deliver even shorter pulses. We use a process dubbed high harmonic generation (HHG), where a laser light pulse of about 4 femtoseconds is converted into a soft X-ray pulse with a duration of about 100 attoseconds. This technology, pioneered by the group of Professor Ferenc Krausz of the Max Planck Institute of Quantum Optics, has been further developed in my laboratory towards spectral and temporal shaping by means of attosecond multilayer mirrors. This allows us to do things such as stretch and compress soft X-ray pulses, shape the temporal and spectral profile, change the polarisation state and eventually focus such pulses in a nanospot.

Could you provide an insight into the development of atto-photoelectron emission microscopy (ATTO-PEEM)?

ATTO-PEEM is an entirely new type of electron microscope, which combines nanoscopic spatial resolution with unprecedented temporal resolution and was developed as a dedicated instrument for our research on ultrafast nanoplasmics.

In ATTO-PEEM, we have combined the photoelectron emission microscope with attoxen soft X-ray pulses from an HHG source. Our vision is to record temporal snapshots of the photoelectrons emitted from metallic nanostructures as a function of space (10 nanometres) and time (100 attoseconds), because these photoelectrons carry information about the lightfield nanolocalisation in the nearfield of the sample structures. However, we are not quite there yet and a big challenge lies in the fact that we can only detect one electron per attosecond pulse, which makes such experiments very cumbersome and time-consuming. With the development of high repetition rate HHG sources and spectral discrimination of the emitted photoelectrons (both aspects successfully employed in our group) I believe we are close to the point where the first time- and space-resolved imaging of nanolocalised plasmonic fields can become a reality.

Which of your laboratory’s discoveries have been of most significance?

We are definitely one of the leading groups worldwide in the development of attosecond optics for spectral and temporal pulse shaping in the soft X-ray range. This technology is a prerequisite for customised attosecond pulses not only in our group, but also serves each attosecond experiment performed in the Krausz laboratory on attosecond physics.

We were one of the first worldwide groups successful in applying such pulses to PEEM, which added another degree of complexity compared to attosecond photoelectron spectroscopy, but also opened the technology towards the investigation of surface-supported nanostructures. With ATTO-PEEM, we are one of the very few laboratories capable of studying photoemissions from nanostructures in four dimensions, space (x and y), time and kinetic energy. This allows us to investigate localised surface plasmon fields, and enables us to study the electronic dynamics of promising new two-dimensional crystalline materials, like graphene or topological insulators, with nanometre and attosecond resolution.
Ultrafast science: pushing back the frontiers of photoelectric physics

Research into the spatiotemporal dynamics of light and matter interactions at the University of Munich seeks to shape the behaviour of materials at the sub-nanometre and attosecond scales

The Complementary Metal-Oxide Semiconductor (CMOS) transistor has been at the heart of developments in modern electronics since the 1970s. Its longevity as a viable solution to information processing needs derives from what is known as Moore’s Law, where it has been possible to pack roughly double the number of these transistors into a chip every 18 months to two years, enabling the progressive development of ever smaller mobile and computing devices with ever greater processing power. However, although CMOS is still the dominant technology for high-performance, low-power and, importantly, low-cost miniaturised devices, it is envisaged that Moore’s Law will not hold for much longer. As a result, switching speeds made possible using CMOS technology will not meet requirements for future applications, so limits will be placed on innovation. The electronics industry is in urgent need of a viable replacement architecture and technology that can deliver much higher switching performance.

In the laboratory of Professor Dr Ulf Kleineberg at the Ludwig Maximilian University of Munich, the possibility of replacing CMOS technology with even smaller transistors capable of extreme performance is being explored using light excitation of surface plasmons in metal nanostructures, especially in terms of inducing the electrons on the surface to respond collectively to the light source in waves. A technology based on this approach would combine what Kleineberg terms ‘the two big advantages of plasmonics’: nanoscale integration and phenomenally fast quasi-optical response – near the speed of light: “If we could steer and control plasmons by the electrical field of an incident light wave, the switching speed compared with conventional electronics could be increased by at least two orders of magnitude into the terahertz regime, more than a trillion cycles per second,” he asserts. However, ways of precisely controlling both the propagation of light pulses and the fast switching and amplification of surface plasmon waves on the nanoscale still need to be found for this prospect to be realised.

Towards Greater Control of Plasmon Nano-Localisation

Specialisation in Kleineberg’s laboratory include nanophotonics, surface and thin film physics and nano-plasmonics, as well as development of advanced instrumentation for observing and measuring electron dynamics in nanostructures. In search of answers, Kleineberg has been investigating means of achieving control of localised surface plasmons in metallic nanostructures made from silver or gold. He has developed a means of fabricating lift-off structures using electron beam lithography in conjunction with a double-layer resist method. For example, in fabricating silver nanostructures, this involves depositing a layer of evaporated silver about 20 nanometres thick onto a nanostructured resist followed by a release step of the ‘unwanted’ material: “The structures work very similarly to radio frequency antennae, concentrating the energy of an electromagnetic wave in a small volume,” he explains. The excitation of surface plasmons by repeated ultrashort femtosecond laser light pulses brings about changes in the optical and electrical properties of the metallic nanostructures – potentially even from insulating to conducting – making it possible to switch between properties very quickly.

Kleineberg has experimented with different shapes of nanostructure, such as bowties, double bowties and ellipsoids, as well as with different...
Technology based on light fields at the nanoscale will ultimately enable ultrafast information processing and will thus play a very important role in the future.

Kleineberg has used carrier envelope phase (CEP) tagging to analyse and control the femtosecond laser light pulses by splitting them into two identical replicas: the first part provides measurements of the electrical field of the pulse, and the second is used in the experiment to examine lightwave/electron interactions on the nanostructure surface under a powerful microscope, named the atto-photoelectron emission microscopy (ATTO-PEEM) device: Kleineberg’s group constructed ATTO-PEEM using the photoelectron emission microscope (PEEM) as a basis. PEEM uses an ultraviolet or soft X-ray source to liberate electrons from a sample surface and then images the relative spatial distribution of the electrons emitted to a resolution limit of about 10 nanometres. The ATTO-PEEM soft X-ray, or extreme ultraviolet source, is based on coherent high harmonic generation (HHG) driven in a gas target by a titanium-sapphire femtosecond laser and can generate individual pulses with attosecond durations in the extreme ultraviolet spectral range. ATTO-PEEM provides four-dimensional microscopy, where the time resolution is achieved from the delay between excitation and probing light pulses, electron kinetic energy by integration of time-of-flight spectrometer and the spatial dimensions – 20 to 100 nanometres – by PEEM. With ATTO-PEEM, Kleineberg has established that the photoemission from metallic nanostructures can be affected by the means of adjusting the durations and frequencies of light pulses. Depending on the configuration of both structure and light pulse, the concentrated energy in the nanostructures can indeed be controlled both temporally and spatially: “If we could optimise and eventually control light field nano-localisation, such nano-antennae could be an integral part of future plasmonic switches or amplifiers,” Kleineberg observes.

Kleineberg is under no illusion that collaboration with world-renowned colleagues is both inevitable in the fast-moving sphere of attosecond physics and fundamental to the success of his experiments. On the experiment, as well as the theory of attosecond physics, and on technological matters such as metrology and sample preparation, he works closely with the Krausz laboratory at the Max Planck Institute of Quantum Optics; on attosecond optics he has a longstanding collaboration with the Centre for X-ray Optics at the Lawrence Berkeley National Laboratory in the US. Kleineberg’s laboratory also participates in two large research consortia: the excellence cluster of the Munich Centre for Advanced Photonics and the German Research Foundation priority programme on ultrafast nano-optics.

For Kleineberg, a key question for the development of feasible plasmonic devices concerns the transfer of energy and momentum from the photon to the electron. How Einstein’s photoelectric effect works is still opaque, especially considering the specific characteristics that nanometre-scale spatial dimensions and ultrafast time scales can confer on materials. While Kleineberg believes that ultrafast information processing based on new nanophotonic devices (e.g. light field-driven electronics or nanoplasmonic devices) is still many years away, and CMOS will prevail for the next decade at least, he also believes that ultrafast information processing and will thus play a very important role in the future, as it already does in long-range optical communication transport: “Our research on the sub-femtosecond dynamics and control of electrons in nanosystems may add a few pieces to this puzzle,” he reveals. To Kleineberg, the main factor that will determine which technology will replace CMOS is time – whichever technology can be quickly leveraged into real-world applications that meet requirements for fast, reliable and powerful performance in tiny devices will be the one that succeeds.