

# TAKING SNAPSHOTS OF ATOMIC MOTION USING ELECTRONS

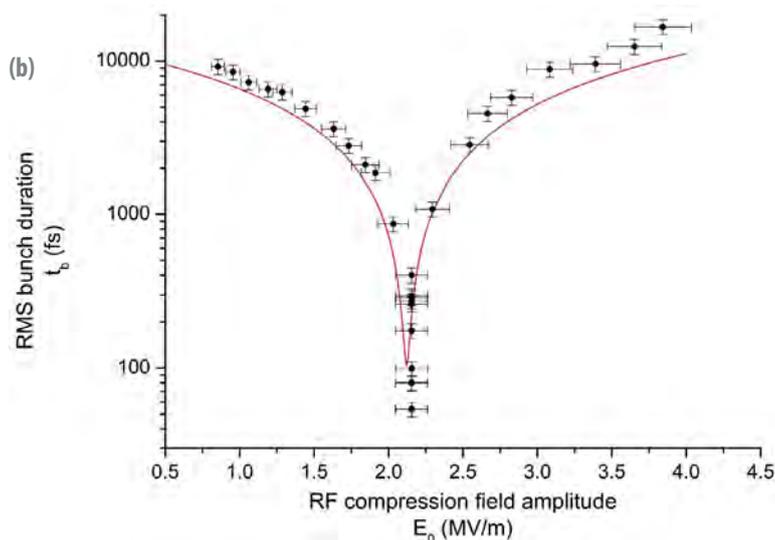
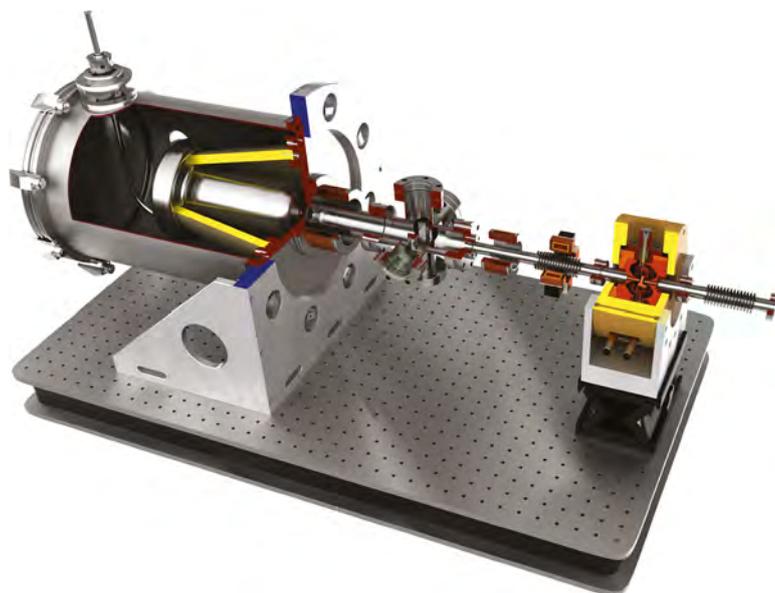
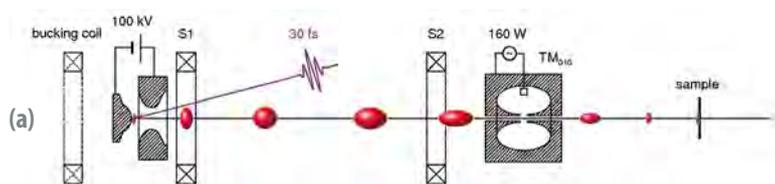
■ **Jom Luiten** – Eindhoven University of Technology, Eindhoven – the Netherlands – DOI: 10.1051/eprn/2015203

55 years after Richard Feynman's famous Caltech lecture 'There is plenty of room at the bottom' [1], heralding the age of nano science and technology, many of the possibilities he envisaged have come true: Using electron microscopy it is nowadays possible to resolve and even identify individual atoms; STM and AFM not only provide us with similar spatial resolution on surfaces, but also allow dragging individual atoms around in a controlled way; X-ray diffraction has revealed the complicated structures of thousands of proteins, giving invaluable insight into the machinery of life.

◀ **P.21:** an artist's impression of the ultracold source in action, with ultracold ions (green) coming out from one side and ultracold electrons (blue) from the other.

**H**owever, despite these spectacular achievements our view on the nano world is still very limited. The reason is that at small length scales things move very fast. The natural time scale of atomic motion in matter, *i.e.*, the vibration period of an atom in a molecule or a lattice, can be as short as 100 fs. This is ten orders of magnitude faster than present-day state-of-the-art imaging techniques can provide, which is video rate at best. Up to now the focus has therefore been almost exclusively on systems in equilibrium, which is just a very tiny sub-class of what is going on down there. Is it possible to actually monitor individual atoms in action, during chemical reactions, macromolecular conformational changes, phase transitions, *etc.*, and thus study structural dynamics at the most fundamental level?

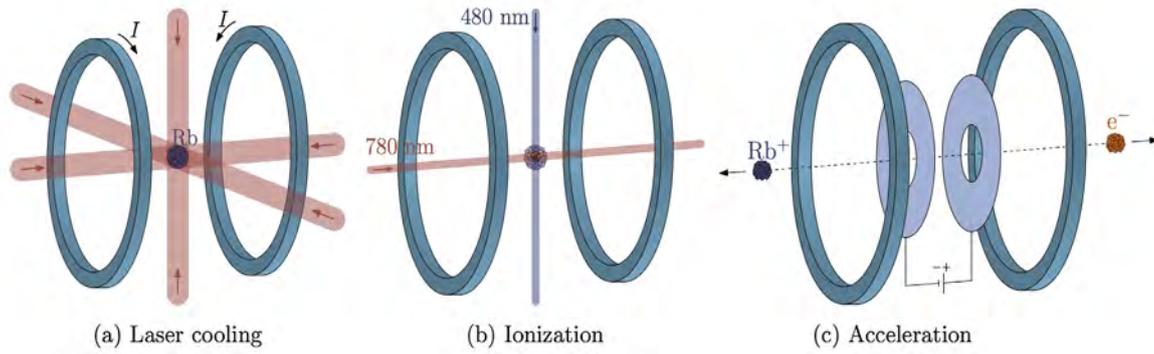
Lasers at optical wavelengths have been providing the required femtosecond temporal resolution for many years already, using the so-called 'pump-probe' method: a process is excited by a femtosecond 'pump' laser pulse, which is subsequently investigated by a 'probe' laser pulse. The pulse pair is exactly synchronized since pump and probe are both derived from the same laser pulse and separated in time by a variable delay line. Using nonlinear optical techniques pump and probe wavelengths can be generated anywhere between the terahertz and soft X-ray regimes. These techniques have proven to be extremely powerful but also have their limitations. First, the shortest probing wavelengths are still much too long for resolving atomic structures; second, light interacts with electrons in matter, so no direct information is obtained on the actual atomic motion, the motion of the nuclei, the lattice, in the process under investigation. Information on the structural dynamics can therefore only be obtained in an indirect, model-dependent way. Ideally one would like to extend femtosecond pump-probe experiments with much shorter wavelengths, *i.e.*, smaller than 1 Å ( $10^{-10}$  m), which implies using either hard X-ray photons or massive particles such as electrons.



### Fast X-ray and electron beams

In the past decade enormous progress has been made in both ultrafast X-ray and electron beam technology. In 2009 the first hard X-ray Free Electron Laser (X-FEL) became operational at Stanford University [2]. The X-FEL delivers nearly fully coherent femtosecond X-ray pulses of sufficient power to record a full diffraction pattern of protein crystals in a single shot, as has recently been demonstrated [2]. Worldwide a few X-FELs are operational now, and several more are under construction. Like synchrotrons, X-FELs are large-scale facilities, but they can accommodate maximally a few users at a time, so beam time is scarce. An essential ingredient for realizing X-FELs was the development of a new generation of ultrabright, femtosecond electron guns. Why not use the electrons produced by these compact and relatively cheap high-brightness guns directly instead of accelerating them to giga-electronvolt energies to generate hard X-ray photons? Thanks to the pioneering efforts of Ahmed Zewail at CalTech and Dwayne Miller at the University of Toronto, in the past decade ultrafast electron diffraction (UED) and ultrafast electron microscopy (UEM) using ultrashort electron pulses generated by femtosecond photoemission have become a serious alternative for X-FELs [3].

◀ **FIG. 1:** (a) Generation of ellipsoidal electron bunches by photoemission with shaped femtosecond laser pulses (left) and subsequent transverse (magnetic coils) and longitudinal (microwave cavity) focussing of the bunches on the target (right); (b) Duration of 100 keV, 0.2 pC electron pulses measured as a function of microwave field strength, demonstrating microwave bunch compression from 10 ps to 100 fs [5].



◀ **FIG. 2:** Schematic of Rb MOT-based ultracold charged particle source: (a) laser cooling and trapping of Rb vapour; (b) two-photon photoionization of Rb atoms; (c) extraction of electron and ion beams from the ionized gas.

## UEM and UED

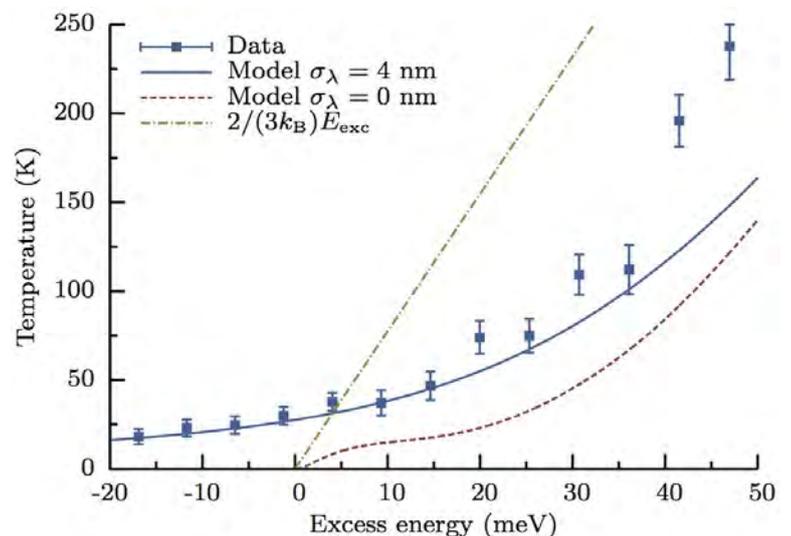
In UEM ultrafast electron imaging is done with megahertz repetition-rate trains of single-electron pulses. Since a full image requires  $10^7 - 10^8$  electrons, UEM can only be used to study highly reproducible samples and processes which can be repeated identically millions of times. In spite of this limitation, many impressive experiments have been performed, mainly on cyclable and robust solid-state systems. Unfortunately, UEM cannot be applied to fragile substances, susceptible to damage by either the electron radiation dose accumulated over the many shots required to record a full image, or by the intense optical fields of the femtosecond ‘pump’ laser pulses. This is particularly true for biological materials, which are extremely sensitive to radiation damage. Moreover, biological processes such as conformational changes of macromolecules, do not follow a single, unique reaction path, and sample preparation methods involved are rarely fully reproducible. For such systems single-shot recording is highly desirable, if not essential. This puts extreme requirements on the intensity and the brightness of beams, which is actually offered by X-FELs. Can this be done with electrons as well?

An alternative to ultrafast electron imaging is ultrafast electron diffraction (UED) of crystalline samples. A high-quality diffraction pattern only requires  $10^6 - 10^7$  electrons and puts less stringent demands on beam coherence. Creating the required ultrashort, highly charged electron pulses is actually not very difficult: simply fire a femtosecond UV laser pulse at a metallic (e.g. copper) cathode surface at a negative potential. Unlike X-ray photons, however, electrons interact very strongly with each other and the cathode surface. This fundamentally limits the maximum achievable coherence. Moreover, packing the required number of electrons in a single femtosecond pulse, inevitably gives rise to a violent Coulomb explosion. This spoils the beam coherence even further and leads to substantial pulse lengthening and thus loss of temporal resolution. A temporal resolution of a few 100 fs can be achieved by limiting the charge in a single pulse to  $10^3 - 10^4$  electrons, so still more than 100 shots are required to record a full diffraction pattern. Fortunately, we found a way to eliminate the problem of the Coulomb explosion.

## Controlling and inverting the Coulomb explosion

Until about a decade ago, the Coulomb problem was considered unsolvable. The only way to ameliorate the problem was slowing down of the Coulomb explosion by accelerating to relativistic speeds as quickly as possible. In 2004, however, we discovered that by proper shaping of the femtosecond photoemission laser pulse, it is possible to create uniformly filled, ellipsoidal electron bunches [4]. These objects, until then considered idealized text book examples, are characterized by linear space charge forces. As a result a uniform ellipsoidal bunch undergoing a Coulomb expansion will remain a uniform ellipsoid, as is illustrated in Fig. 1a. The linearized Coulomb expansion can be inverted by linear external fields provided by charged particle optics. The transverse expansion can be handled straightforwardly using regular charged particle lenses, but inversion of the longitudinal expansion requires a temporal lens, which is not a standard optical element. In 2010 we demonstrated how a compact, low-power 3 GHz microwave cavity can be used to invert the longitudinal

▼ **FIG. 3:** Effective source temperature vs. excess photon energy using femtosecond photoionization of a Rb MOT-based electron source [6]; green dash-dotted curve: effective temperature assuming a simple plasma model and equipartition; red dashed curve: exact classical model prediction for monochromatic photoionization; blue solid curve: exact classical model prediction convolved with femtosecond laser bandwidth; blue squares: experimental data.



expansion and compress bunches containing more than  $10^6$  electrons to 100 fs bunch lengths [5], see Fig. 1b. The resulting pulsed beam is of sufficient quality to record full diffraction patterns of simple atomic crystals in a single 100 fs shot. This approach has been adopted in the last few years by several of the leading UED groups [3]. UED has developed into a powerful technique for visualizing ultrafast structural dynamics in condensed matter systems. The beam coherence is however not yet sufficient for diffraction of complicated macromolecular structures due to the limitations on the initial coherence set by traditional photocathodes.

### The cathode problem

Electrons near a metal surface are pulled back due to image charge forces. To simultaneously extract  $10^7$  electrons from a micron-sized area requires electric fields of  $\sim 10^{11}$  V/m, two orders of magnitude higher than the highest field strengths that can be attained without vacuum breakdown. The obvious way out is to increase the surface area to tens of microns. Unfortunately, this goes at the expense of beam coherence, which scales inversely

proportionally with source size. A compact and proper way to quantify the coherence or beam quality of an accelerated bunch of particles is the phase space density

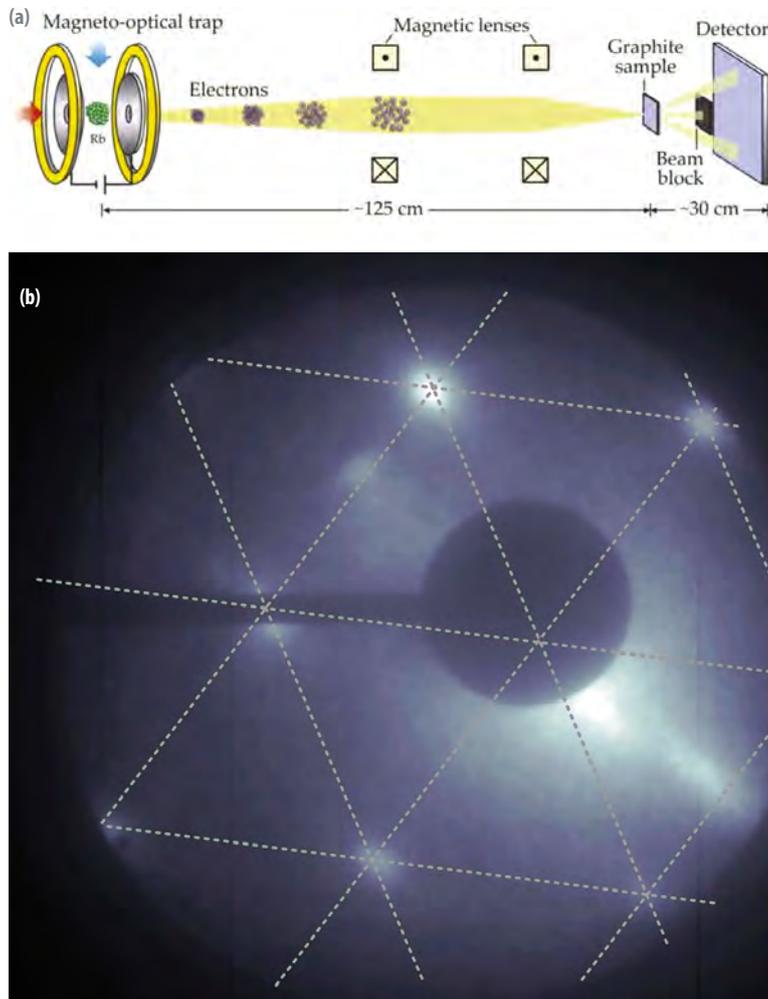
$$f = n \cdot \lambda_{th}^3,$$

where  $n$  is the number density and  $\lambda_{th} = h/\sqrt{2\pi mkT}$  the thermal De Broglie wavelength associated with the average thermal motion of the particles in the bunch, characterized by an effective temperature  $T$ , with  $h$  Planck's constant, and  $m$  the particle mass. The phase space density is also proportional to beam brightness. In optical terms,  $\lambda_{th}$  is the equivalent of the (spatial or temporal) coherence length. Due to their fermionic nature,  $f=1$  is the maximum attainable phase space density for electrons. For a conventional metallic photocathode  $kT \approx 0.5$  eV, *i.e.*,  $T \approx 5000$  K. Combined with a density of  $10^7$  electrons in a bunch a few tens of microns across in all three dimensions, this results in a phase space density  $f \approx 10^{-7}$ . Clearly there is lots of room for improvement. The traditional approach is to improve coherence by increasing the density, reaching for the ultimate ideal of a point source. In electron microscopy sources the best that has been achieved in this way is  $f \approx 10^{-3}$ , with, however, only a single electron at a time. We have developed an alternative approach, which relies on a drastic reduction of the source temperature, allowing for much larger source sizes, and thus much more charge per pulse, while retaining a high degree of coherence.

### Ultracold electron source

Our ultracold source is based on near-threshold photo-ionization of a laser-cooled and trapped atomic rubidium vapour (see Fig. 2). Using standard techniques, rubidium atoms are cooled to sub-millikelvin temperatures and trapped in a magneto-optical trap (MOT). Using two-photon ionization, employing a continuous 780 nm laser to populate the  $5P_{3/2}$  state and a tunable 480 nm femtosecond laser to ionize the excited atoms, electrons can be extracted from the cold atoms with very little kinetic energy. Recently, we have demonstrated the generation of picosecond electron bunches with  $T \approx 10$  K from a source 30 micron in size [6]. Figure 3 shows the effective source temperature  $T$ , measured as a function of the excess energy with which the electrons are liberated in the photo-ionization process (see Fig. 3). The data agree very well with the theoretical curve, which is based on a classical description of the electron emission from a single, motionless rubidium atom. Recently, the first diffraction patterns have been produced with a 10 keV beam from the ultracold source, which confirm the independently measured coherence properties of the source (see Fig. 4 and [7]). These experiments were done with a few hundred electrons per bunch, to prevent Coulomb interaction from spoiling the coherence.

▼ FIG. 4: Graphite electron diffraction using an ultracold source. (a) Schematic of experimental setup [7]; (b) Diffraction pattern recorded with a 10.8 keV electron beam. The width of the diffraction peaks is consistent with the independently measured effective source temperature  $T = 10$  K [7].



The achieved phase space density  $f \approx 10^{-7}$  is comparable to state-of-the-art photocathode guns. If we were able to stash  $10^6$  electrons in the same bunch, *i.e.*,  $10^3 - 10^4$  times higher density, resulting in a phase space density  $f \approx 10^{-4} - 10^{-3}$ , then 100 fs, single-shot electron diffraction of complicated macromolecular crystals, and maybe even single-shot imaging come within reach. Is this possible?

### Future

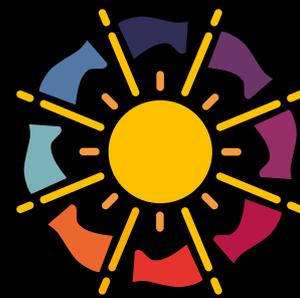
To achieve this goal we plan to combine the ultracold electron source with bunch-shaping and bunch-compression techniques. Recently, the group of Rob Scholten of the University of Melbourne demonstrated how a spatial light modulator can be used to shape the excitation laser profile and thus the initial spatial distribution of ultracold electron bunches extracted from a MOT into almost any desired 2D form [8]. To actually realize uniform ellipsoidal electron bunches, the initial distribution needs to be shaped in 3D, which means that both the excitation and ionization laser profiles need to be controlled. In addition, the effect of the ion background on the electron distribution during bunch extraction has to be dealt with, making bunch shaping a greater challenge than in the case of a flat photocathode. Once this last obstacle has been taken ultrafast, single-shot electron diffraction of protein crystals, and possibly even ultrafast, single-shot electron imaging, will become a reality. ■

### About the author

**Jom Luiten** obtained his PhD in 1993 at the University of Amsterdam. After working for several years on superconducting X-ray detectors and spending a year in industry he turned to accelerator and beam physics at Eindhoven University of Technology (TU/e) in 1998. Currently he is heading the TU/e group Coherence and Quantum Technology, conducting research on coherent charged particle beams, ultracold plasmas, ultrafast electron diffraction and microscopy, and coherent light-matter interaction.

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