ATTOSECOND SCIENCE AND TECHNOLOGY

by H. Niikura, F. Legaré, J. Itatani, M.Y. Ivanov, D.M. Villeneuve and P.B. Corkum

Laser pulse durations fell continuously (Fig. 1) from the early days of the laser until 1986, reaching 6 fs, just 3 periods of 600 nm light [1]. The underlying reason for this advance was the demands of science for ever-faster measurements. Each advance in lasers opened new phenomena for measurement. Each new measurement pointed out the importance of developing better ultrafast sources.

The momentum towards ever shorter pulses continued until 6 fs pulses were reached. These pulses only contained 3 periods of the carrier frequency (600 nm light). For progress to continue a new technology based on much shorter wavelength carrier frequency was needed. Developing this new "attosecond technology" took about 15 years, although the basic outline of the technology was clear about 10 years ago [2,3]. Attosecond technology relies on femtosecond lasers but is otherwise a major departure from the technology that preceded it.

At the core of attosecond technology is a strong laser field that exerts control over a continuum electron wave packet. A laser pulse ionizes an atom or molecules, thereby transferring an electronic wave packet into the continuum near the peak of the laser field.

OVERVIEW OF ATTOSECOND TECHNOLOGY

At the core of attosecond technology is a strong laser field that exerts control over a continuum electron wave packet. The underlying process is quite simple. A laser pulse ionizes an atom or molecule, thereby transferring an electronic wave packet into the continuum near the peak of the laser field. Ionization is the first step in the 3-step quasi-static model of strong field (attosecond) science [4].

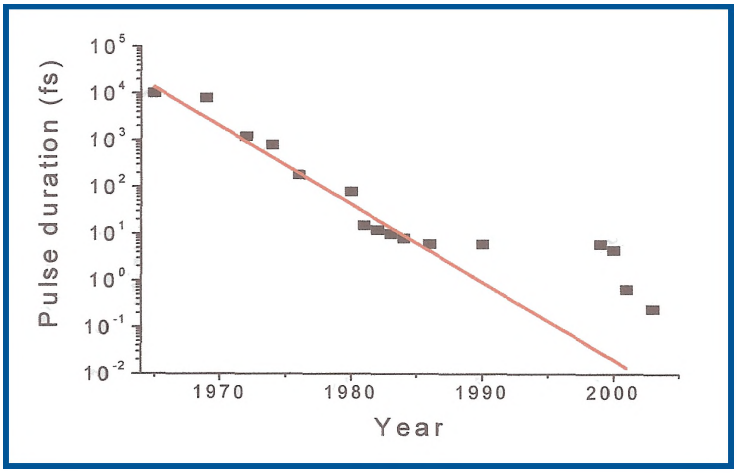

Fig. 1 **The pulse duration of ultrashort pulse lasers since shortly after lasers were discoverred.**

This electron wave packet plays a central role in attosecond technology. For high ionization potential atoms and infrared radiation (800 nm or longer) DC tunneling models [5] describe the birth of the electron wave packet. In this quasi-static description, time is used as a parameter to describe the time variation of the laser field. Since the electron wave packet is born by ionization of a well-defined initial state by a coherent process, the electronic wave packet is fully coherent with the state from which it departed. Tunnel ionization is illustrated in Fig. 2.

Once free, the electron wave packet is accelerated in the laser field and dispersed by it. The parts of the wave packet that are produced first experience the field for a longer time and never return to the parent atom/ion.

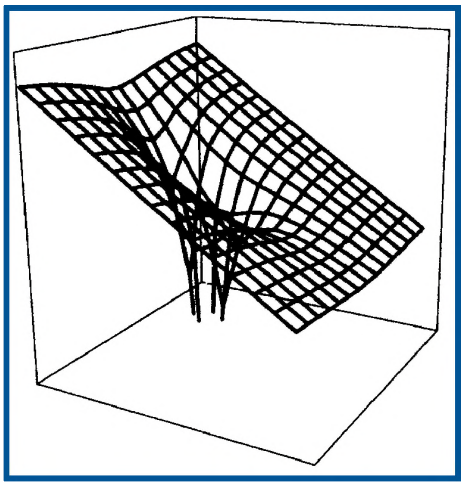

Fig. 2 **The instantaneous potential seen by a bound atomic electron at a given instant in the laser field. For high ionization potential atoms, such as rare gases, the electron tunnels from this potential almost as if the field were a static field.**

The parts of the wave packet formed following the crest of the field are driven back. Some collide (usually called a recollision, emphasizing that the electron returns to its parent) with high kinetic energy. Propagation of the electronic wave packet in response to the strong laser field is the second step in the quasi-static 3-step model.

Electron motion in a laser field is completely coherent.

Hiromichi Niikura, F. Legaré [1], J. Itatani [2], M.Yu Ivanov, D.M. Villeneuve and P.B. Corkum <paul.corkum@nrc.ca>, National Research Council of Canada,Ottawa, Ontario;
[1] Ph. D. student at the University of Sherbrooke;
[2] Postdoctoral Fellow at University of Ottawa.

Thus the electron retains its coherence with its parent wave function from birth to re-collision.

In spite of the classical language that is commonly used in strong field science, an electron wave packet is not a classical particle. Classical physics is a useful aid to our intuition and it offers some quantitative predictions. Some quantum mechanical features can be added "by hand". For example the electron tunnels through the lip of the potential barrier shaped a bit like the lip of a tipped cup. Heisenberg's uncertainty principle requires that the electron has an uncertain momentum given by $\Delta x \Delta p = h$ [5, 6]. This is also true in the longitudinal direction. In this direction it is not as well understood, but is thought to be approximately the same magnitude [7].

It is the final step of the 3-step process that makes attosecond technology such a powerful technology. During the re-collision, the electron wave packets can inelastically interact with its parent ion, exciting or multiply ionizing it [8,9]. It can elastically scatter from the ion, producing a diffractive or holographic pattern of the ion [10-12]. Finally the re-colliding electron wave packet can interfere with the remainder of the wave packet that did not ionize, as shown in Fig. 3, inducing an oscillating dipole that produces high frequency light [13]. In this process the kinetic energy of the electron is converted into photon energy producing high frequency radiation.

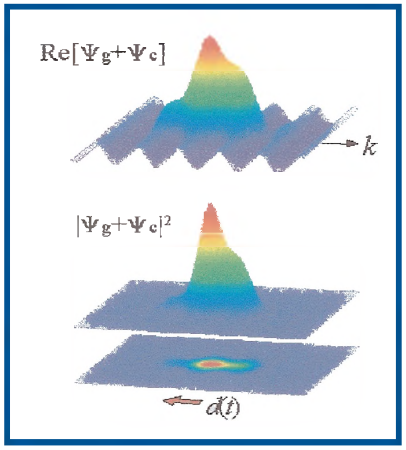

Fig. 3 A snapshot of the induced dipole

To summarize, directed by the *strong coherent laser field*, a *coherent electron wave packet* is created and interacts with its *parent atom/molecule*. During this interaction which lasts less a fraction of an optical period it can produce *coherent photons*. These coherent electrons and photons are the focus of most attosecond science.

A CLOSER LOOK AT THE RE-COLLISION ELECTRON

The laser field naturally confines the electron re-collision to a fraction of the laser period following the wave packet formation. Figure 4 characterizes the re-collision from the perspective of its re-collision partner during the few cycles following its ionization. Normally quantum mechanics would forbid us to know exactly when ionization occurred. Later, when we discuss experiment to measure the electron dynamics we will have to specify a method of measurement that allows it to be measured.

Figure 4 shows the time structure of the re-collision. Different processes have different cross-sections and so, although only one electron is involved, it is instructive to adopt the language of electron beams. Figure 4 shows that an external beam would need ~ 10^{11} A/cm^2 to reach a similar probability of collision and it would have to be confined to a series of peaks with the predominant peak arriving with sub-femtosecond precision. Such current densities are unavailable from any conventional source. Not shown it the kinetic energy of the electrons. Concentrating on the major peak, it starts at zero kinetic energy at early times and increases to 3.17 U_p at the time of peak current density (~1.7 fs, near the time that the laser electric field crosses zero) and then falls to zero again before the first satellite pulse arrive. It is quite common with 800 nm light to have U_p ~ 50 eV. Here U_p is the ponderomotive energy, $U_p = q^2E^2/(4m\omega^2)$ where q and m are the electronic charge and mass; E and ω are the laser peak electric field and frequency. If infrared radiation is used, U_p can reach 1 keV [14]. (Note, this the same ponderomotive energy that plays such a prominent role in plasma physics.)

The part of the wave packet that causes the kinetic energy to sweep from low (~0.8 fs) to high energy (~1.7 fs) arises from electrons born well after the peak of the laser field and are known as short trajectory electrons. The part of the wave packet that sweeps back down in kinetic energy results from electrons that are born just after a field crest and followed a long trajectory, re-colliding late in the optical cycle.

The second and following peaks correspond to electrons that miss on the first attempt, but are "Coulomb focussed" to re-collide on the second or subsequent attempt. As their trajectories become more and more complex the probability decreases. Phase matching ensures that these late re-collisions do not play a large role in harmonic generation but can be important in inelastic scattering.

We have confirmed that the current density in Fig. 4 is correct through a correlated measurement on H_2 [11]. The measurement exploits double ionization of H_2. We measure photofragment ions to obtain information of the current density as well as the dynamics of non-sequential double ionization. This measurement represents the fastest real-time measurement of electronic dynamics that has been reported.

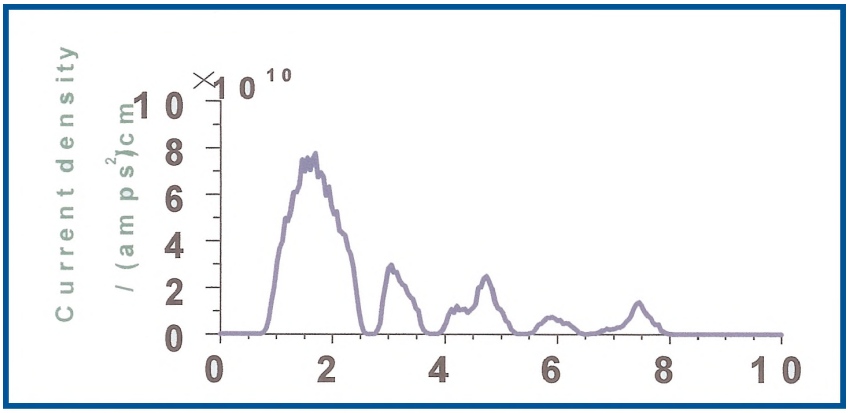

Fig. 4 The time history (measured in femtoseconds) of the electron-ion re-collision.

A correlated measurement is unusual in ultrafast measurements. With our 40 fs pulse, quantum mechanics does not allow us to know when ionization will occur. However, since we are using H_2 we know that whenever ionization does occur, it creates two wave packets at once — an electron wave packet that we wish to measure and a vibrational wave packet on H_2^+. Our measurement of the current density exploits correlation. Correlated decay in the presence of a strong laser field may offer a completely new method for attosecond measurement [15]. It may allow us to make ultrafast measurements in areas of science where real time measurements were previously impossible, such as the atomic nucleus [16].

PRODUCING ATTOSECOND OPTICAL PULSES

Attosecond optical pulse formation is not a correlated processes. The electronic wave packet beats with that portion of the atomic or molecular wave function (Fig. 3) remaining in the initial state that is ionizing, producing a photon (*i.e.* it recombines to the original state). Since ionization can occur during any ½ cycle of the laser field the recombination can also occur over many ½ cycles. All that is required is for a new part of the initial wave function to be ionized during the previous crest of the laser field and for there to be population remaining in the ground state at the time of re-collision. Therefore, with a long pulse, each ½ period there is a burst of XUV radiation resulting in a frequency comb in XUV wavelength spaced by twice the photon energy of the laser field. The duration of the pulses should be ~ 1 fs and each individual pulse should chirp from low frequency to high frequency and then back down just as the individual electron pulses do (Fig. 4).

Single attosecond pulse formation involves isolating one of these XUV bursts. High harmonic generation is equivalent to forming a train of attosecond optical pulses [13, 17] — the Fourier transform of a broad bandwidth series of XUV bursts is a broad sequence of harmonics. The pulses that make up the harmonics are separated from each other by ½ laser period (or 1.3 fs for an 800 nm laser pulse). The question is "how can we select a single pulse from the train of pulses"?

There are two ways to select a single attosecond pulse. Either a few cycle pulse can be used [18] or a pulse with time dependent polarization [2]. Preferably the pulse would be carrier envelope phase controlled [19] so that there is only one peak of the laser field that is capable of produce a high kinetic energy re-collision. Currently, phase controlled ~6 fs pulses produce the best single (isolated) attosecond pulses [20]. Fig. 5 shows the electron re-collision probability obtained using a 7 fs pulse.

Attosecond pulse trains have been produced in many labs around the world for many years. However, only recently has it been possible to measure the pulses. Thus the central issue in attosecond science for the past decade has been measurement. Measuring attosecond pulses is closely related to their application to time-resolved measurement.

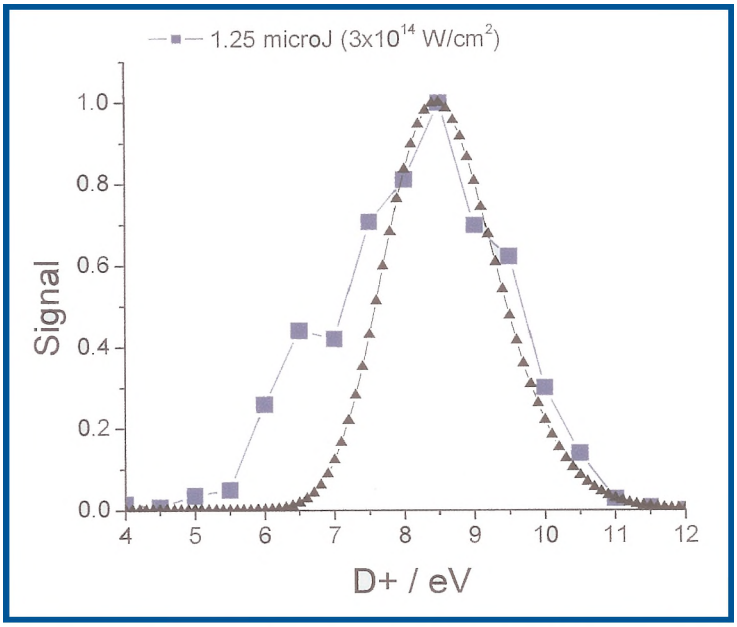

Fig. 5 Current density experienced by a D_2^+ following ionization by a 7-fs duration, $3x10^{14}$ Wcm^{-2}, 800 nm pulse. In this experiment correlated D^+ fragments are measured. The triangles plot the signal that is expected from the first re-collision. The data points show that there is only a small contribution from subsequent re-collisions. This would produce a nearly isolated attosecond optical pulse.

MEASURING ATTOSECOND PULSES

For the XUV pulses the answer is that we employ the same basic technology for production and measurement. Fundamentally attosecond science exploits attosecond electron wave packets controlled by strong laser fields. To measure the pulse we first form an electron wave packet replica of the XUV pulse and then we measure it. This procedure is similar to that used in optical streak cameras.

There are a number of ways that the electron replica can be measured [21 22, 17], but we will concentrate on the attosecond streak camera. It was proposed very early [3, 13] and it is used in all single attosecond pulse measurements. It has also been used to measure Auger decay dynamics of core holes in Krypton [23].

The attosecond streak camera uses an atom as a photocathode and the time dependent laser field to label the time of ionization. More specifically, an electron bound to an atom is not free to respond to a laser field. However, once an attosecond pulse ionizes it, the field can accelerate it. The energy the electron gains from the field is added to that energy that it acquired from the XUV pulse. Photoelectrons formed at the beginning of the attosecond pulse gain a different energy from the laser field than those formed later. Thus, by measuring the electron energy spectrum, we gain enough information to fully reconstruct the optical pulse, both its duration and its chirp [21, 23].

To review up to this point, attosecond technology can deliver electron pulses to an atomic or molecular target that has sub-femtosecond duration and sub-Angstrom wavelength. Pulse durations of electrons can be measured to ~ 1 fs precision and the duration of XUV pulses can be measured to a precision of ~ 70 attoseconds [22] using the attosecond streak camera. The same principle can be used to construct spectral shearing interferometry for electron wave packets. This method is analogous to spectral phase interferometry for direct electric field re-construction (SPIDER) used for characterizing ultrashort visible pulses. There is no obvious limit using attosecond SPIDER [24].

It is widely perceived that the development of attosecond pulses is one of the most important developments of science during the past few years [25, 26]. This perception is largely an article of faith. In the introduction we commented on the symbiosis between ultrafast technology and its scientific applications. So, is the faith justified? What important new science is possible because of attosecond technology? While it is too early to know for sure we will end this article with very short sketches of three of the possible new directions.

OBSERVING MOLECULES

Attosecond science allows time resolved measurements to be made with attosecond resolution. Thus, we will be able to measure the fastest electronic processes involving valence electrons. Equally, because the electron wavelength is so small it will allow molecular structure with high spatial precision so, attosecond science is really attosecond/ Angstrom science. Until recently such powerful technology for studying atoms and molecules seemed unrealistically far in the future, if they could ever be produced at all. However, it is no longer the case. Attosecond technology, especially attosecond electrons, offers exactly this combination. Already the first steps have been made along this path [15].

OBSERVING AND CONTROLLING ELECTRONS

Electrons are critically important. They form the bonds that hold molecule and solids together. Making and breaking of bonds is the very essence of chemistry. The natural time scale of electrons is attoseconds. If we wish to create, observe or control electron wave packets, attosecond technology is needed. Already the first steps towards imaging electrons, both their amplitude and their phase, have been made [27].

ELECTRON-ELECTRON CORRELATIONS

Electrons repel each other. This is true in atoms and molecules as it is elsewhere. However, theories of atomic structure are largely based on a single electron approximation, treating the other electrons as an average background. How electrons arrange and how they collectively respond to a stimulus cannot be observed now except under special circumstances such as in Rydberg atoms. Attosecond technology may give us an experimental tool for observing electron-electron interaction in normal matter. The first steps along this path are the real time measurements of Auger decay in krypton [23] and experiments on correlated double ionization [28].

Attosecond science is still emerging — and so different from the optical science that preceded it — that it is unlikely that we understand more than the most vague outline of its significance. There is room for creativity and new ideas from anyone who reads this article.

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