

Chapter 1

Attosecond Science

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Abstract Attosecond technology builds on, and contributes to, important historic directions in science. For this reason, the science has the depth to yield important discoveries for a long time. Unfortunately the title “attosecond” science biases us to think mostly about dynamics but many of the most important applications of “attosecond technology” may not be related to dynamics at all. This broad set of applications point to the future impact of the technology.

It is a pleasure to introduce this new publication with some thoughts of the past and future of attosecond science. During the last decade we have engineered a radical advance in Quantum Electronics. As Fig. 1.1 shows, the duration of the shortest pulse that we can create has decreased by more than one order-of-magnitude after about a decade in which there had been little advance. We have achieved this by moving to shorter wavelength as also illustrated in Fig. 1.1. Therefore, you will see that we developed not only the shortest pulses but also the shortest wavelength sources available with laser-based methods. In fact, as you read this new volume you will see that there are four scientific traditions from which attosecond physics grows and these traditions open four complimentary ways for attosecond science to influence the world’s scientific agenda.

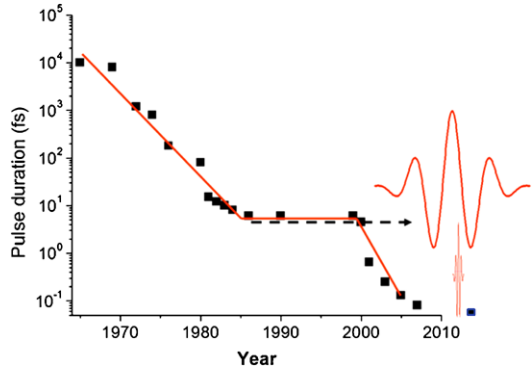
We can understand one tradition by thinking of attosecond science as the current frontier of ultrafast science—a 50 year old sub-field of laser science, with even deeper roots. For 50 years, ultrafast science has been a major direction of research. It has retained its freshness because time is a natural frontier of all of the physical sciences. As we pursue time to shorter and shorter intervals, we inevitably find unexplored science, opening new processes for inspection. From the perspective of where we sit today, we are a very long way from any natural boundary. Therefore, for the foreseeable future we can expect technology to drive the production of even shorter pulses, motivated by the demand to study ever faster phenomena.

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Fig. 1.1 The shortest pulse that could be generated as a function of time since mode locking was developed



So attosecond science inherits the benefits of 50 years of research into concepts and approaches for fast measurement. As attosecond researchers, it is our task to improve, adapt and apply those methods. From our current perspective, just 10 years into the “attosecond age”, there remains a lot of work to do. Because the sources are still not intense enough, we cannot directly apply attosecond pump-attosecond probe spectroscopy—to say nothing of the more powerful femtosecond methods such as 2-D spectroscopy [1]. However, we have also added something new. We have learned to use a low frequency (visible in our case) pulse as either the pump [2, 3] or probe [4] or both [5]. With a low frequency pump or probe, time resolution can arise through the many photon nature of the absorption [5] or through a coherent interaction [4]. With this mixed approach, we have made major advances. For example, using the “attosecond streak camera” [6, 7], we can time resolve the appearance of electrons from different bands in materials after attosecond pulse illumination [4]. We can also time resolve cascading recombination in atomic media (to mention but two of the recent advances [8]). This mixed approach may find natural applications in ultrafast terahertz spectroscopy.

Nonlinear optics is the second tradition from which attosecond science springs. Nonlinear optics is also a 50 year old sub-field of optical science. In fact, all ultrafast measurements are *of necessity* nonlinear. Since nonlinear optics is so fundamental to time resolved measurements, we should expect the development of non-perturbative nonlinear optics to open new measurement paradigms. In fact, this expectation is true. It allows attosecond pulses to be measured as they are being formed [9, 10]. Furthermore, non-perturbative nonlinear optics allows us to excite and observe unexpected phenomena. For example, we can time resolve tunnelling [11] or excite attosecond bound-state electronic wave packets in atoms [5, 12] or molecules [2] launched by tunnelling. We can image molecular orbital wavefunctions [13, 14], something that chemists are taught is conceptually impossible. New time-resolved spectroscopes are being developed that exploits this new nonlinearity [15–17].

The form of non-perturbative nonlinear optics that has led to attosecond pulse generation has a clear physical model—the “recollision” model [18, 19]. The success of the model is based on the control that a short, intense pulse can exert on an ionizing electron. The classical intuition that this model encourages has been very

helpful. It suggests that control can go much deeper—to weakly bound electrons and perhaps even more strongly bound electrons. The better we control laser fields, the better we will control these electrons and through electrons, matter.

“Recollision” emphasizes a third tradition on which attosecond science can draw—one of the oldest fields of physics—collision physics. It is through collisions that science first learned about the structure of matter, from the structure of the atom, to the structure of the nucleus, to the structure of the proton. Therefore, through recollision, optics gains systematic access to this kind of structural information with spatial resolution appropriate to the collision energy range in which recollision operates. This includes atomic and molecular structure [20–22] and it will eventually include nuclear structure [23].

But it is not all a one-way street. Attosecond science offers two important tools to collision physics that were previously lacking. First, it offers the opportunity to time-resolve collision events [24, 25], something that was not systematically available previously. This may be particularly helpful for ultrafast studies of the dynamics in the atomic nucleus [23]. Secondly, since collisions can be timed with respect to optical pulses, collision physics gains access to the powerful pump-probe methods of optics. A pump pulse might initiate dynamics to be studied by delayed collisions or it might control the system—aligning or orienting molecules, for example, to simplify scattering.

The collision-inspired aspect of attosecond science offers us a very powerful dream. It should be possible to simultaneously make spatial and temporal measurements with spatial resolution on the interatomic (or even nuclear) scale and time resolution on the valence electron time scale [25, 26] or even, ultimately on the nuclear time scale.

Finally, the mathematics of short pulse formation and the technology of their generation require that attosecond pulses lie in the XUV or soft X-ray spectral region. This is the spectral region that synchrotrons opened for study about 30 years ago. Thus we can benefit from the 30 years of synchrotron experience. Using atomic specific resonances, and other related X-ray methods, we will be able to probe molecules or solids.

Of these grand traditions, attosecond science is not the only radically new “game-in-town”. Just as femtosecond technology has given birth to attosecond technology, so also synchrotrons have given birth to free-electron lasers [27]. While each of these technical advances is important, when viewed together, it is clear that we are experiencing a historic advance in photonics technology whose ramifications we are only beginning to explore. One major task we have for the next decade is to step outside of our own field and identify those areas of science where we can have a major impact. There are many sub-fields to explore.

I would like to take this opportunity to very briefly highlight one specific new direction that work in my laboratory is opening—polar molecules [28–30], although there is no attosecond dynamics involved—in fact no dynamics at all except for the dynamics we impose by recollision. However, as I have argued, our tools can be applied much more broadly than to only study dynamics. If we can provide new insight into polar molecules—a class of molecules that are very important to chemistry—that is an equally important accomplishment.

The Issue Chemistry is highly directional. When reagents approach, they experience local fields that modify the collision and therefore the reaction. When at least one of the reagents is a polar molecule, then the reaction proceeds differently depending on the direction of approach of the reagents. This is the field of stereo chemistry. Can we offer new insight into polar molecules?

A re-collision electron is a natural and highly sensitive tool to study these local fields:

- It is charged.
- It is launched by directional tunnelling and it ends with a re-collision from the same direction.
- It can report on the recollision electron characteristics through the attosecond photons that it emits.
- We have powerful tools for measuring both the amplitude and the phase of the electron or the associated photons, allowing us to compare different sides of the molecule.

From these studies we will gain a detailed understanding of an electron tunnelling from each side of a molecule and also the local field structure around the molecule.

The Technology It is now feasible to orient polar molecules in a robust manner. Those interested in the approach are referred elsewhere [29, 30].

In oriented polar molecules, on alternate half-cycles of the laser field, the electron tunnels from one side of the molecule or the other. The subsequent motion of this electron wave packet is initially likewise restricted to one side for both the short trajectory electrons and the long trajectory electrons. Thus, both the ATI electrons and the attosecond pulse that they launch encode the natural asymmetry of the orbital, the Stark shift and the local electronic environment through which the electron departs. It is this asymmetry that is also responsible for stereo chemistry.

If the driving pulse were to gate an isolated attosecond pulse, then, as we change the carrier-envelope phase (CEP), we would change the re-collision direction and, in fact, the whole time structure of the attosecond pulse. The latter is the case, because the frequency dependent transition moment that describes recombination is different in both amplitude and phase for an electron recolliding from opposite directions [28].

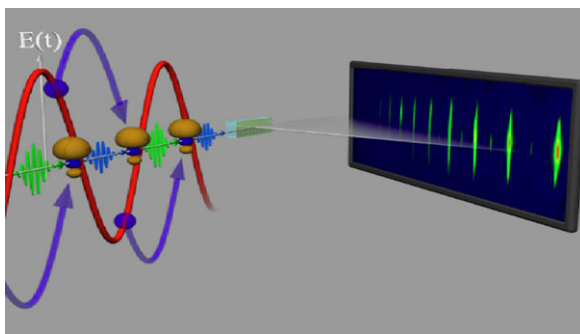
If the driving pulse has multiple cycles, then the interference between subsequent attosecond pulses in the train create even harmonics as illustrated in Fig. 1.2 as the weak spectral lines between the stronger high harmonics (taken from [28]).

While these are early experiments we will soon be able to bring the full array of attosecond tools to bear on polar molecules.

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Fig. 1.2 Illustration of high harmonic generation from CO molecules (taken from [28]). The asymmetry of CO leads to even harmonics as seen in the experimental spectrum that stretches from H15 *on the right* to H29 *on the left* are oriented



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