Nobel Lecture: The route to attosecond pulses*

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CONTENTS

The history of attosecond science can be divided into four periods:

(1) Background and discovery of high-order harmonics (1987–1992)
(2) Progress in technology and understanding (1993–2000)
(3) Characterization of attosecond pulses and first applications (2001–2011)
(4) Opening to different research fields (2011–2023)

I will go through the first two parts and leave the third part to my colaureates Pierre Agostini and Ferenc Krausz. In Sec. IV, I will just say a few words about some recent applications.

I. BACKGROUND AND DISCOVERY OF HIGH-ORDER HARMONICS

The field of attosecond science would not have existed without the invention of the laser in 1960 by Theodor Maiman[1] (Maiman, 1960). The properties of the laser that are important in our research in attosecond science are the generation of short laser pulses and the ability to achieve a high intensity by focusing the laser light. The invention of the laser opened up new research fields. The first one is nonlinear optics[2] where new frequencies are generated when laser light is focused into a material. In the first experiment performed by Peter Franken and his colleagues in 1961 (Franken *et al.*, 1961), a ruby laser was focused into a crystal of quartz, and the second harmonic of the laser was produced. In this process, the material absorbs two photons with frequency ω (the laser frequency) and emits one photon at the double frequency (2ω).

Another research field that emerged after the invention of the laser is "atoms in strong fields" or the study of multiphoton processes, which were predicted theoretically already in 1931 by Maria Göppert-Mayer (Göppert-Mayer, 1931). Figure 1 gives some examples of multiphoton processes, starting with two-photon excitation, multiphoton ionization (Voronov and Delone, 1966), above-threshold ionization, where an atom is ionized by absorbing more photons that are necessary for ionization (Agostini *et al.*, 1979), and multiple ionization, which was the focus of my thesis (L'Huillier *et al.*, 1983).

A natural extension of this work was to detect the fluorescence that could be emitted from excited atoms or ions. In 1987, at the Commissariat à l'Énergie Atomique, Saclay, France, we set up an experiment to observe this fluorescent light. We used a Nd:YAG laser, with a wavelength of 1 μm and a pulse duration of 40 ps. Since fluorescence is emitted in all directions, we had two axes of detection, along the direction of the laser propagation axis and perpendicular to it. Figure 2 is a photograph of this setup. The red lines indicate both the laser propagation axis, coming from the right, and the two detection axes. The intensity achieved in the medium was higher than 10^{13} W/cm^2. The medium was provided by a pulsed gas jet, delivering an atomic density of the order of 100 mbar. This was the time before personal computers and the data were recorded by a printer, coupled to the detector.

We never saw light in the perpendicular direction and we did not see much fluorescence light. We saw, instead,

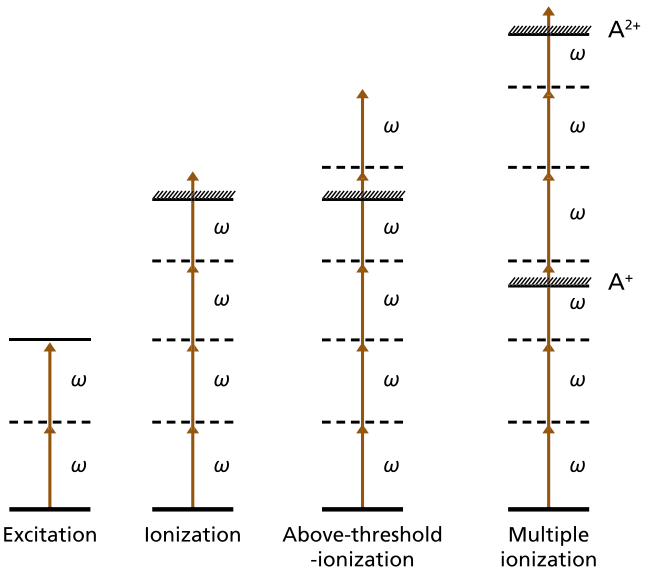

FIG. 1. Diagrams representing some multiphoton processes.

*The 2023 Nobel Prize for Physics was shared by Pierre Agostini, Ferenc Krausz, and Anne L'Huillier. This paper is the text of the address given in conjunction with the award.

[1]A related Nobel prize was awarded in 1964 to C. H. Townes, N. G. Basov, and A. M. Prokhorov "for fundamental work in the field of quantum electronics, which has led to the construction of oscillators and amplifiers based on the maser-laser principle."

[2]Half of the 1981 Nobel Prize was awarded to N. Bloembergen and A. L. Schawlow "for their contribution to the development of laser spectroscopy." In particular, Bloembergen developed spectroscopy methods based on nonlinear optics. The other half of the prize went to K. Siegbahn for "his contribution to the development of high-resolution electron spectroscopy."

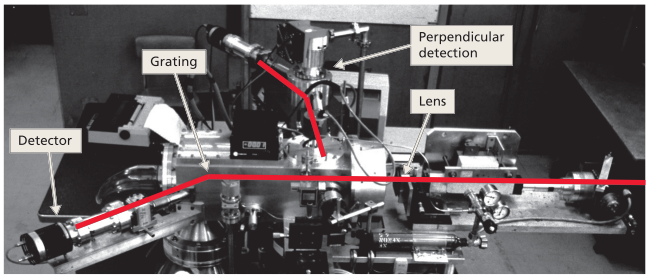

FIG. 2. Photograph of the experimental setup. The laser is coming from the right. The data were recorded on a printer shown on the left. Courtesy of Lidyl, CEA.

high-order harmonics of the laser light (McPherson *et al.*, 1987; Ferray *et al.*, 1988). Figure 3 shows one of the first spectra obtained in argon. The peaks correspond to odd-order harmonics, with a frequency equal to an odd multiple of the laser frequency, from the 13th to the 31st.

The intensity distribution of these high harmonics is represented in Fig. 4. We observed a typical behavior, with a decrease for the first orders, followed by a plateau, where all the harmonics have the same intensity, and ending up with a cutoff at the 31st order. This result was very unexpected! We did not anticipate seeing so many high-order harmonics, with comparable intensities. The polarization (P) induced in a medium by the interaction with an electromagnetic field (E) is often expanded as

$$P = a_1 E + a_3 E^3 + a_5 E^5 + \cdots.$$

This expression requires that the first order is much larger than the third, that the third order is much larger than the fifth, etc. This was not the case. Only odd orders are produced due to inversion symmetry, which kills even orders. The radiation produced by high-order harmonic generation (HHG) in gases is in the extreme ultraviolet region of the spectrum, with photon energies of several tens of eV.

High-order harmonic spectra resemble the distribution of overtones produced by some musical instruments, for example, the alto violin. In both cases, there is a strong interaction, between a laser field and atoms, or between a bow and a string,

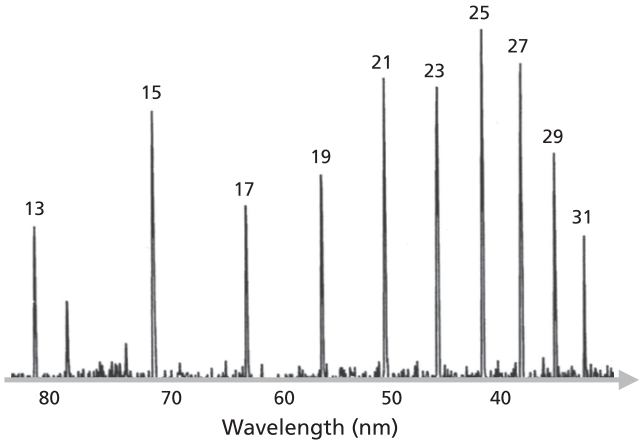

FIG. 3. Harmonic spectrum obtained in argon.

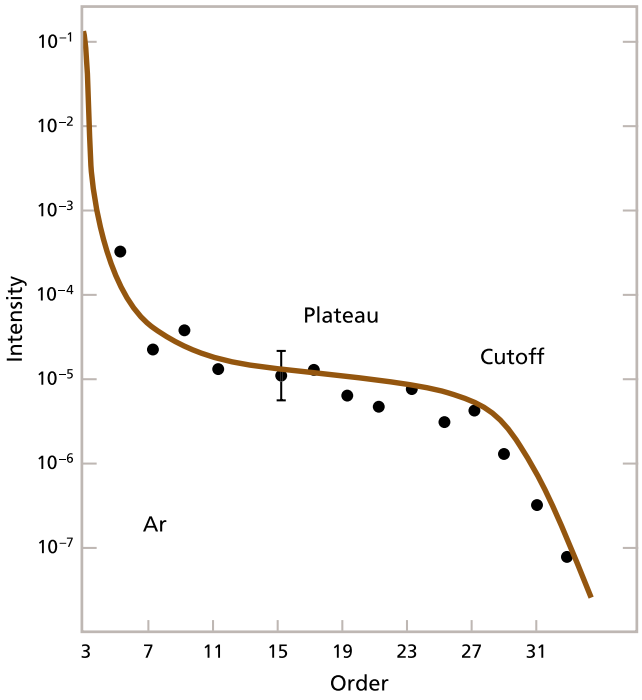

FIG. 4. Harmonic distribution in argon. Adapted from Ferray *et al.*, 1988.

resulting in many overtones. Of course, in the latter case, the overtones are those of the initial sound (tone) frequency, while the high-order harmonics discussed in this lecture are those of the laser light.

The physics of high-order harmonic generation includes both atomic physics and nonlinear optics. One needs to calculate the response of an atom to a strong radiation field, i.e., to solve the time-dependent Schrödinger equation (TDSE),

$$i\hbar\frac{\partial\Psi}{\partial t} = -\frac{\hbar^2}{2m}\nabla^2\Psi + [V(r) + eE(t)\cdot r]\Psi,$$

where Ψ is the time-dependent wave function, $E(t)$ is the laser field, and $V(r)$ is the atomic potential. The laser-atom interaction, comparable to the atomic potential, is here described within the dipole approximation. One also needs to consider the response of all the atoms in the medium, a problem traditionally described by Maxwell's equations of electromagnetism, which can be simplified to a wave equation,

$$\nabla^2 E - \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = \frac{1}{\epsilon_0 c^2}\frac{\partial^2 P}{\partial t^2},$$

where E is the generated field and P the nonlinear polarization induced by the laser field. Physically, two conditions are required for high-order harmonic generation: each atom needs to generate these harmonics and for each harmonic frequency, the light generated by all of the atoms in the medium should add in phase, which is called phase matching. Continuing the musical analogy, the ensemble of atoms constitutes a gigantic orchestra of trillions of musicians, which need to play at the same pace.

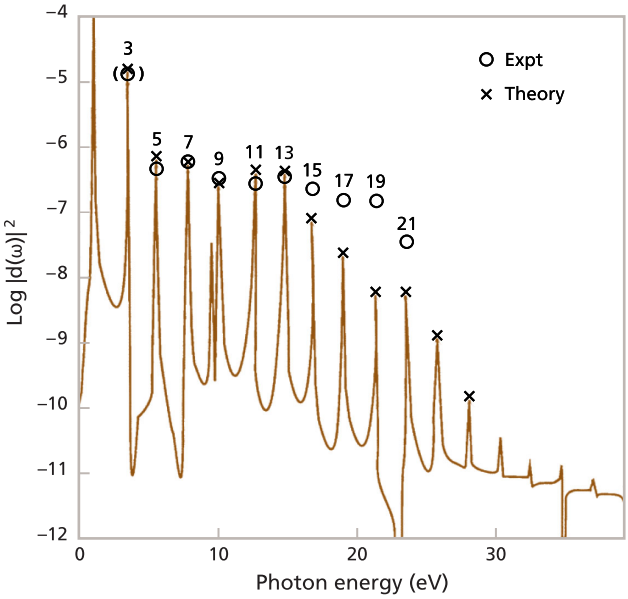

FIG. 5. Comparison between experimental data [circles, from Ferray *et al.* (1988)] and numerical results obtained by solving the TDSE. Adapted from Kulander and Shore, 1989.

The first attempt to describe HHG was simply to solve these two equations numerically. Figure 5 presents a comparison between numerical simulations of the single atomic response of xenon atoms (Kulander and Shore, 1989) with the experimental results obtained in xenon (Ferray *et al.*, 1988). The agreement between theory and experiment is quite good. This result was in itself surprising since phase matching was expected to be more and more difficult to achieve as the process order increased. Numerical simulations (L'Huillier, Schafer, and Kulander, 1991) combining the solution of the TDSE and of a wave equation describing the generation of harmonics in a macroscopic medium showed that this was not the case. The plateau and cutoff behavior of high-order harmonic generation experimentally observed could be numerically reproduced. This did not mean that the physics behind it was understood, however.

During this same period, the question was raised whether the harmonics were temporally synchronized, or phase-locked, which would lead in the time domain to a train of short, attosecond, pulses (Farkas and Toth, 1992; Harris, Macklin, and Hänsch, 1993). Phase locking means that the maxima of the harmonic waves all coincide at a certain time. Due to the periodicity of the interaction, after a half-laser period, the minima will be at the same time. After another half-laser period, the maxima will again be at the same time, etc. If the harmonics are phase-locked, they interfere constructively every half-laser cycle, leading to the emission of a train of light pulses, with a duration of the order of 100 as, i.e., 10^{-16} s. The more harmonics are added in phase, the shorter the pulse duration.

The interest of HHG in the production of short light pulses was motivated by the fact that the evolution of the pulse duration of lasers had come to a limit of a few femtoseconds (Fork, Greene, and Shank, 1981; Dietel, Fontaine, and Diels, 1983; Fork *et al.*, 1987). The reason for this limit, also called the "femtosecond barrier" is that the duration of one cycle of visible laser light is typically a couple of femtoseconds. For example, a cycle of titanium-sapphire laser with a wavelength in the near-infrared is 2.6 fs. Light pulses should contain at least an optical cycle. Obtaining a light pulse below a femtosecond requires a frequency in the ultraviolet or extreme ultraviolet range. The second condition to produce a short light pulse is that the bandwidth should be large. The light generated in the interaction of atoms with a strong laser field possessed both properties: a central frequency in the extreme ultraviolet range and a large bandwidth. This motivated us to investigate the question of the phase locking of the high harmonics, as well as to find methods for the measurements of short attosecond light pulses.

II. PROGRESS IN TECHNOLOGY AND UNDERSTANDING

At the beginning of the 1990s, progress in high-power laser technology improved the experimental conditions for HHG, allowing the generation of high photon energies at a high repetition rate. The chirped pulse amplification was discovered and implemented in many laboratories (Strickland and Mourou, 1985). A new laser material was found, titanium sapphire (Moulton, 1986). A new method for the generation of short laser pulses, Kerr-lens mode-locking, was discovered (Spence, Kean, and Sibbett, 1991). At Lund University, Sune Svanberg, Anders Persson, and Claes-Göran Wahlström invested in this new technology and bought the first amplified titanium-sapphire laser system in Europe.

In parallel to this progress in laser technology, dedicated instruments to study high-order harmonic generation were designed and built. For the first time, personal computers could be used and acquisition programs were written, allowing us to process data in a completely different way (L'Huillier and Balcou, 1993). My first two Ph.D. students, Philippe Balcou and Pascal Salières, and I were invited to perform joint experiments in Lund, combining our dedicated instrument and the new femtosecond terawatt titanium-sapphire laser (Wahlström *et al.*, 1993). The combination of a high repetition rate (10 Hz) and dedicated instrumentation including automatic data acquisition allowed us to study HHG as a function of different parameters, for example, the laser intensity (see results in Fig. 6). The titanium-sapphire laser rapidly became the standard laser to generate high-order harmonics (Macklin, Kmetec, and Gordon, 1993).

At approximately the same time (1993), a real breakthrough was made in the theoretical understanding of high-order harmonic generation with the three-step model, proposed by Kenneth Kulander and Kenneth Schafer (Kulander, Schafer, and Krause, 1993; Schafer *et al.*, 1993) and independently by Paul Corkum (Corkum, 1993). Just a few months later, Maciej Lewenstein proposed a quantum formulation of this model, called the strong field approximation, which would have an enormous impact on the field (Lewenstein *et al.*, 1994). The three-step model is illustrated in Fig. 7. In an intense laser field, the atomic potential is strongly distorted, and at a certain time during the laser cycle, an electron in the ground state may tunnel through the potential barrier formed on one side of this distorted potential.

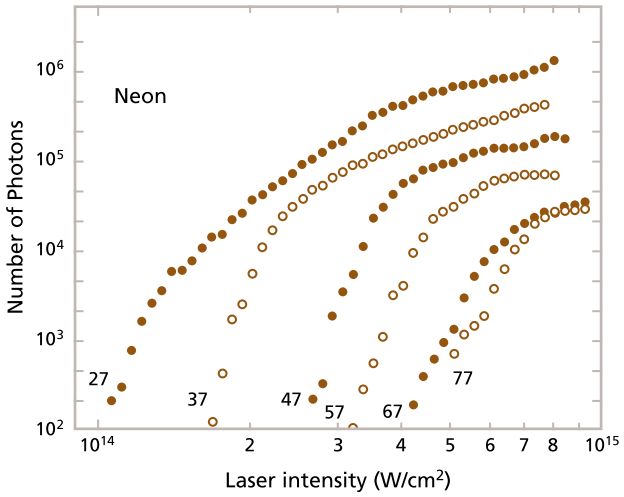

FIG. 6. Variation of the harmonic yield in neon for harmonics 27, 37, 47, 57, 67, and 77. Adapted from Wahlström *et al.*, 1993.

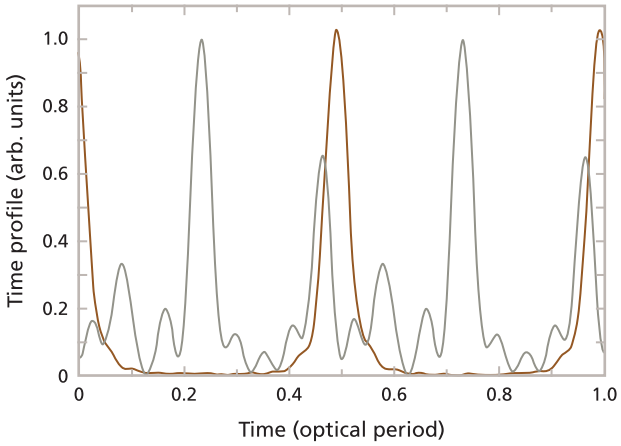

FIG. 8. Light emission during an optical cycle. The gray line corresponds to the single-atom response while the brown line is a result obtained by also including the propagation in the nonlinear medium. Adapted from Antoine, L'Huillier, and Lewenstein, 1996.

The liberated electron is driven away until the laser electric field changes sign. The electron is then pushed back toward the atomic core, and there is a certain probability of recombination back to the ground state. Along its trajectory, the electron gains kinetic energy, and the excess energy is emitted in the form of a high-energy photon. Considering all of the possible electron trajectories, an electron wave packet and subsequently a light pulse are created with a short duration, high frequency, and broad bandwidth. This process is fully coherent and is repeated every half-laser cycle. The repetition every half-laser cycle, with a change of sign between consecutive light pulses, leads in the frequency domain to a spectrum of high-order harmonics.

In Fig. 8, we show the temporal emission from an atom during a laser cycle, in gray (Antoine, L'Huillier, and Lewenstein, 1996). In contrast to the short pulses expected if all of the harmonics are phase-locked, the calculation shows a complex temporal structure with several pulses per half-laser cycle. Two main pulses can be identified due to two types of possible electron trajectories, short or long, leading to the same photon energy. The better understanding brought by the three-step model and the strong field approximation also provided a more complex picture, where the generation of short attosecond light pulses once every half-laser cycle was not clear.

The two trajectories were observed in 1998, in an experiment performed in Lund together with Marco Bellini and Theodor Hänsch (Nobel Prize laureate 2005) within the framework of a European collaboration. The experiment consisted of investigating the coherence properties of high-order harmonics (Bellini *et al.*, 1998). The laser beam was split into two beams, which were focused at two different positions, perpendicular to the laser propagation axis. The harmonic beams generated from the two sources, which acted as a double slit, were observed in the far field (see Fig. 9). A spectrometer allowed us to separate the different harmonic components. The interference pattern observed in Fig. 9(a) indicates that the generated light is spatially coherent. One can also observe two contributions to the harmonic emission: a collimated beam, due to the short trajectory and a more divergent part, which is attributed to the contribution of the long trajectory. In Fig. 9(b), we delayed the emission from the two sources by 15 fs. The interference pattern is still present for the collimated radiation while it has disappeared for the more divergent contribution, indicating a shorter

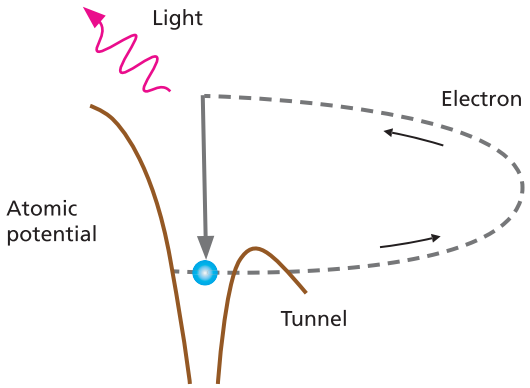

FIG. 7. Illustration of the three-step model.

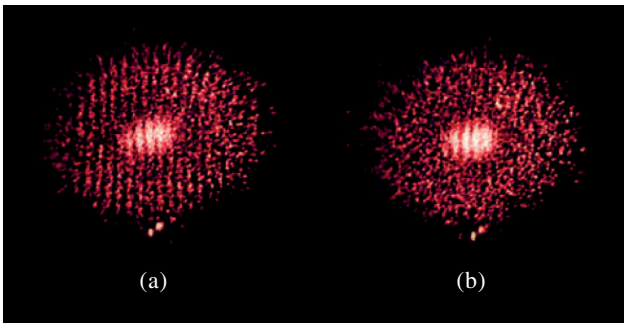

FIG. 9. Interference fringe pattern produced by two harmonic sources, acting as a double slit. On the left, the two sources are synchronized, while on the right they are delayed by 15 fs. The harmonic order is 15, the generation medium is argon. Adapted from Bellini *et al.*, 1998.

coherence time. The two trajectory contributions have different properties, which allows us to manipulate them and possibly select one of them. In the example shown in Fig. 9, a simple way to eliminate most of the long trajectory contribution is to aperture the beam in the far field. This would be used by Pierre Agostini and co-workers in his seminal measurement of attosecond pulses in a train (Paul *et al.*, 2001), based on an interferometric technique suggested by Véniard, Taïeb, and Maquet (1996).

During the 1990s, progress was also made in the understanding of phase matching in the nonlinear medium. Phase matching requires that the phase mismatch between the generated harmonic field and the nonlinear polarization driving it is equal to zero. It can be written as

$$\Delta k = \Delta k_{\mathrm{at}} + \Delta k_{\mathrm{foc}} + \Delta k_{\mathrm{traj}},$$

where Δk_{at} is the phase mismatch due to dispersion in the partially ionized nonlinear medium, Δk_{foc} comes from focusing, and Δk_{traj} is due to the accumulated phase by the electron on its trajectory. The last term is dependent on the trajectory and therefore, in some conditions, the two contributions to the harmonic emission are not simultaneously phase-matched. This was confirmed by numerical simulations, indicated by the brown line in Fig. 8. In this case, only one attosecond pulse is emitted per half-laser cycle and the harmonics are almost phase-locked (Antoine, L'Huillier, and Lewenstein, 1996). At the end of the 1990s, it became progressively clear that attosecond pulses could be generated in the interaction of atoms with a strong laser field, but it was extremely important to verify this result and experimentally measure attosecond pulses.

In parallel to this discussion about the existence of attosecond pulses, a lot of progress was achieved both in ultrafast laser and in HHG technology. Let me briefly mention some of these results. Few-cycle lasers became available, following progress in postcompression techniques (Nisoli *et al.*, 1997). To use these lasers for HHG or other applications, the carrier-envelope phase, which is the phase offset between the laser electric field and the pulse envelope, had to be measured and controlled (Telle *et al.*, 1999). This was going to be very important for the generation of reproducible isolated attosecond pulses (Baltuška *et al.*, 2003; Goulielmakis *et al.*, 2004). Novel medium geometries like cells or capillaries were explored (Rundquist *et al.*, 1998). Molecules were used to generate high-order harmonics (Fraser *et al.*, 1995). This would lead to a completely new branch in attosecond science, called attosecond spectroscopy (Niikura *et al.*, 2003). Higher photon energies were produced using short-pulse lasers and especially long-wavelength drivers (Sheehy *et al.*, 1999). A few years later, photon energies above 1 keV could be obtained (Popmintchev *et al.*, 2012). Finally, the energy per pulse was optimized, using long focusing geometries and reaching microjoules (Takahashi, Nabekawa, and Midorikawa, 2002).

III. CHARACTERIZATION OF ATTOSECOND PULSES AND FIRST APPLICATIONS

The first measurements of the duration of attosecond pulses, either in a train (Paul *et al.*, 2001) or individually (Hentschel *et al.*, 2001) will be presented by my colaureates Pierre Agostini and Ferenc Krausz. They will also describe some applications of attosecond pulses.

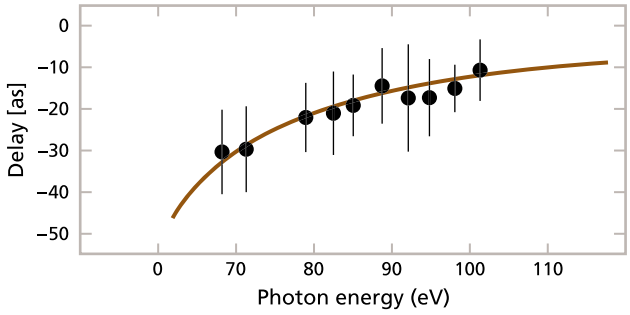

FIG. 10. Difference in measured photoionization time delays between the $2s$ and $2p$ subshells in neon atoms. Adapted from Isinger *et al.*, 2017.

IV. OPENING TO DIFFERENT RESEARCH FIELDS

I will finish this presentation by giving you two examples of applications. One is basic research while the second is much more applied.

One of the simplest examples of electron dynamics is photoionization, where one electron is removed from an atom after the absorption of a high-energy photon.[3] Thanks to attosecond pulses, we can now measure how long it takes for an electron to propagate in the material (Cavalieri *et al.*, 2007) or in the atomic potential (Schultze *et al.*, 2010; Klünder *et al.*, 2011). In case of atoms, these tiny time intervals, usually of the order of tens of attoseconds, have been called photoionization time delays.

I will not describe here the details of the measurement procedure, but use again an analogy from music. A conductor keeps the pace by making a wave motion with his or her hands. This is also the case for the tip of a metronome. Similarly, we measure oscillations in photoelectron spectra and compare the position of the maxima for different processes or photon energies to get temporal information. In the case of neon photoionized simultaneously in both the $2s$ and $2p$ subshells by an attosecond pulse train, in the presence of an infrared probe field, our measurements allow us to determine differences in photoionization time delays of a few tens of attoseconds, as shown in Fig. 10. As explained by Isinger *et al.* (2017), in this particular case, this difference can be attributed essentially to the influence of the probe infrared field used for the measurement.

The second example concerns applications of high-order harmonic generation for imaging and microscopy (Fuchs *et al.*, 2017; Gardner *et al.*, 2017). Modern lithography techniques, based on illumination at 13.5 nm, and reaching structure dimensions less than 10 nm, should be accompanied by metrology techniques using radiation sources with high

[3]This phenomenon was explained theoretically by Albert Einstein in 1905. He was awarded the Nobel Prize in Physics "for his services to theoretical physics and especially for his discovery of the law of the photoelectric effect."

brightness, high spatial coherence, broad bandwidth, and a central wavelength in the extreme ultraviolet range, such as HHG radiation (Porter *et al.*, 2023).

ACKNOWLEDGMENTS

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