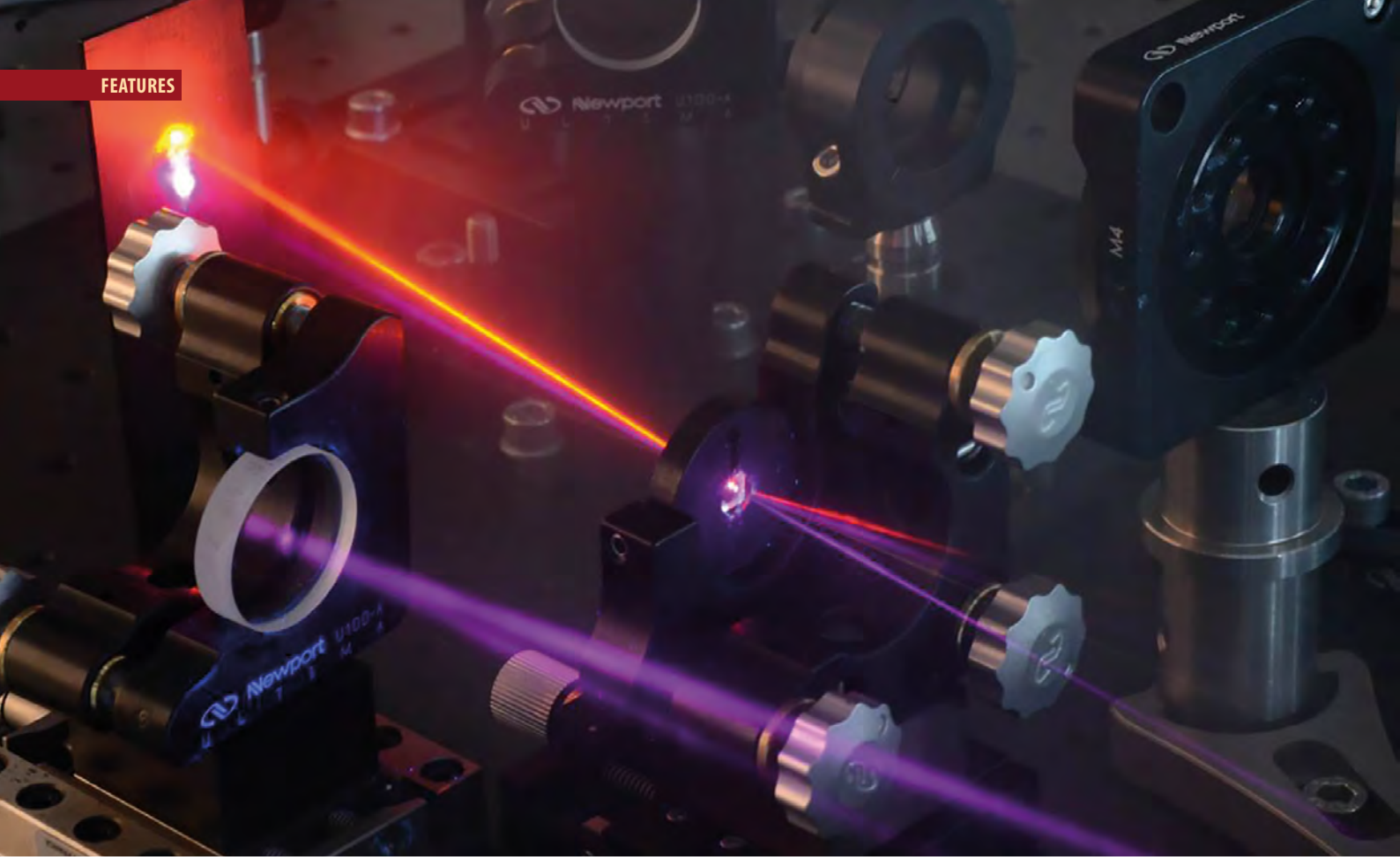

A NOBEL PRIZE FOR ATTOSECOND PHYSICS BASED ON EXTREME NONLINEAR OPTICS

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DOI: https://doi.org/10.1051/epn/2024102

Nobel Prizes related to lasers are awarded for their application in pioneering research areas, as was the case in 2023. Lasers are closely linked to 13-14 Physics Prizes, involving new discoveries, inventions, or research methods. The list is long, including optical fibers, optical tweezers, frequency combs, femtochemistry research, and research related to trapped particles. Lasers also play a crucial role in detecting gravitational waves and in holography. The 2023 award fits into this powerful series. The Prize and the oeuvre of Pierre Agostini, Ferenc Krausz and Anne L'Huillier shows how state-of-the-art laser technology enabled the emergence of extreme nonlinear optics and attophysics and, in turn, how attosecond science triggered the development of revolutionary light sources that are now used in medical diagnostics research or the semiconductor industry.

Until 1960s, the word 'optics' appeared only once in the short citations for Nobel Prizes. However, following the invention of lasers in 1960, the development of related research fields was uninterrupted, enabling the emergence of new fields in optical science. The 1964 Prize was awarded for the development of the "maser-laser principle" and the most recent Prize for laser development, awarded to Gérard Mourou and Donna Strickland in 2018, was for a new optical amplification principle, which plays a pivotal role in femtosecond laser systems. Chirped pulse amplifiers (CPA) proved to be indispensable in contemporary laser technology.

In this article, we present the work of the 2023 Physics Laureates from the perspective of nonlinear optics and extreme nonlinear optics. For this, we need to start with some basics. From Maxwell's equations, in a non-magnetic, charge-free isotropic medium, we can derive the following wave equation with a polarization source term:

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

where **P** denotes the polarization vector, **E** the electric field strength vector, ϵ_0 the dielectric constant, and c the speed of light. Additionally, the relationship $\mathbf{P} = \epsilon_0 \underline{\underline{\chi}}\mathbf{E}$ also holds, where χ is the susceptibility, which in the most general case is a tensor quantity. Thus, it is evident that the electric field of light propagating in a medium induces polarization, which in turn acts as a source of the electromagnetic field. When the induced polarization linearly depends on the electric field, we are in the realm of linear optics. This effect explains basic optical phenomena, such as the refractive index of media or birefringence.

Higher-order effects can occur when the induced polarization is in a nonlinear relationship with the electric field. In such cases, the following relationship is valid:

$$\mathbf{P} = \sum_i \mathbf{P}^{(i)} = \underbrace{\epsilon_0 \underline{\chi}^{(1)}\mathbf{E}}_{\text{linear term}} + \underbrace{\epsilon_0 \underline{\chi}^{(2)}\mathbf{EE} + \epsilon_0 \underline{\chi}^{(3)}\mathbf{EEE} + \ldots}_{\text{nonlinear terms}} \qquad (2)$$

Let's not dwell on the linear susceptibility $\chi^{(1)}$, responsible for the refractive index, but instead focus on the nonlinear components of polarization. The second-order susceptibility $\chi^{(2)}$ is responsible for several interesting nonlinear optical phenomena, such as second-harmonic generation: in a suitable crystal, red laser light can be converted into blue or ultraviolet light (introduction figure). Similarly, the Pockels effect is an example of a $\chi^{(2)}$-effect: an external DC field can change the refractive index of a crystal. The third-order nonlinear susceptibility $\chi^{(3)}$ is also important in a laser lab, as it relates to the dependence of a medium's refractive index on laser intensity, with implications such as the self-focusing of an intense laser beam, for example.

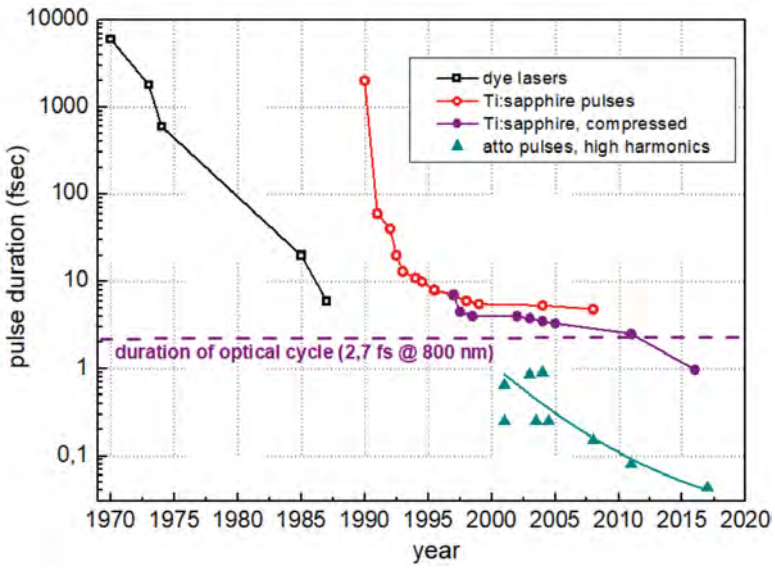
Next, let's estimate the electric field strength at which nonlinear effects become observable. At a distance corresponding to the first Bohr radius of a hydrogen atom, the internal electric field strength within the atom is approximately 5×10^{11} V/m. Suppose that nonlinear effects are observable at three orders of magnitude lower field strength, around 10^8 V/m - this aligns well with laboratory observations. To achieve a peak intensity of 10^8 V/m in the focal spot of a pulsed laser beam, we need to reach a peak intensity of 1.3×10^9 W/cm², based on the relationship $I = \frac{c\epsilon_0}{2} E^2$. This light intensity can be achieved by focusing a 10 W continuous laser beam onto a roughly 1 µm² area. Since lasers are needed to achieve such intensity or electric field strength in the focal spot, optical second-harmonic generation was first observed in 1961 by Peter Franken and his colleagues, just a year after the advent of lasers [1].

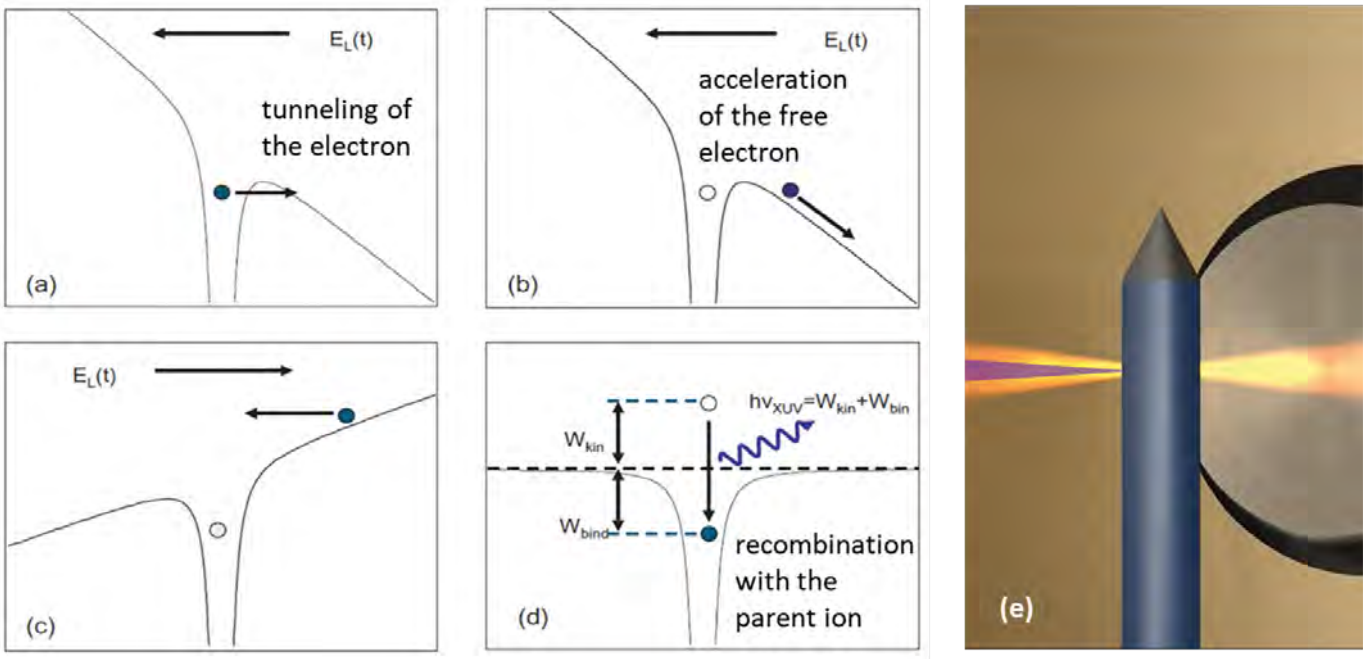
Nonlinear optics has since played a significant role in numerous applications, primarily by enabling the efficient generation of laser light of different colours and the switching of light within the beam based on the Pockels effect. However, as the numerical example above shows, in classical nonlinear optics, the external laser field is still only a perturbation of the internal atomic fields. Therefore, this realm of light-matter interaction is often referred to as perturbative nonlinear optics.

Attosecond sources involve interactions with higher laser intensities, where the field of the laser illuminating the medium can no longer be considered a perturbation of the atomic fields but is comparable to them. Thanks to the development of lasers, such intense light has been achievable since the 1970s. Higher laser intensities can be attained in two ways: by amplifying the optical pulses of lasers or by compressing them in • • •

◂ **P.16 introduction picture:** Frequency conversion in a nonlinear crystal (copyright @Reinhard Kienberger)

▾ **FIG. 1:** Change in the pulse length of lasers providing the shortest pulses, on a logarithmic scale, supplemented with the duration of attosecond pulses. The explosive development of titanium-sapphire laser technology in the 1990s is evident. The figure also marks the duration of the optical cycle of 800nm laser light. The spectrum of pulses shorter than this becomes almost unmanageably broad, covering the entire visible range.





▲ **FIG. 2:** Generation of high-order harmonics – the fundamental process of extreme nonlinear optics. **(a)** The most weakly bound electron of the atom exits via tunnel ionization if the intense laser field sufficiently distorts the potential. **(b) (c)** The free electron is accelerated by the oscillating laser field, during which it gains significant additional energy. **d)** Due to the oscillating field, the electron returns to the ion, with which it recombines, releasing its excess energy as a high-energy extreme ultraviolet (XUV) photon. **(e)** Generation of attosecond pulses in a gas jet. The laser pulse coming from the right drills a hole in the metal cap, and the XUV radiation generated on the noble gas atoms in the cap exits in the same direction as the laser, with less divergence, after the extreme nonlinear optical interaction.

• • • time. Figure 1 illustrates the timeline of producing increasingly shorter pulses. For simplicity, until the 1980s we only plotted dye lasers that provided the shortest pulses up to that time. Researchers during that time also used solid-state and excimer lasers for light-matter interaction experiments with longer pulses. The 1990s was then marked by the emergence of a revolutionary new technology, that of the titanium-doped sapphire (Ti:sapphire) lasers. Thanks to the advancement of Ti:sapphire technology and chirped-pulse-amplified, high-intensity laser chains, the investigation of extreme interaction phenomena gained new momentum.

Let's now look at what extreme nonlinear optics is and the interactions necessary for generating attosecond pulses. The definition of extreme nonlinearity is when the external laser field can not be considered as a perturbation to the field binding the electrons in atoms. Interesting phenomena take place when the laser field becomes comparable to the atomic field strength. Starting from the numerical example related to the hydrogen atom mentioned earlier, this occurs at a laser intensity of 3×10^{16} W/cm^2, and laboratory experience is that around 10^{15}-10^{16} W/cm^2 extreme nonlinearities take place. In such cases, the oscillating field of the laser light distorts the atomic potential to such an extent that the most weakly bound electron leaves the atom via tunnel ionization. After ionization, the electron gains additional energy in the electric field of the laser pulse and returns to the parent ion in the case of linearly polarized light. It can then recombine with the ion, releasing the excess energy gained during its motion in the laser field as high-frequency (extreme ultraviolet, XUV) light: harmonics of the original laser frequency, potentially hundreds of orders higher, can appear depending on the intensity and wavelength. This process is shown in Figure 2. High-harmonic generation with this mechanism was first observed around 1987-1988 in the USA and in France and the pioneering work of Anne L'Huillier [2] and others in this field [3,4] involved both the experimental investigation and the theoretical understanding of this process.

Harmonics are – in other words – a periodic modulation of the spectrum, here up to the XUV regime. In the time domain this means a modulation, too – in other words: a train of pulses. This is clear from Fourier theory and was understood pretty soon [5] after the first measurements of the well modulated HHG spectra. The challenge in the 1990s was to measure these pulse trains. Techniques well known from the femtosecond regime, using an ultrafast pulse, generating a replica and measuring the pulse "by itself" failed. These autocorrelation techniques rely on the nonlinearity of a medium as described above. Unfortunately, these nonlinearities decay strongly with the use of shorter wavelengths. Autocorrelations of (X)UV pulses was only demonstrated for lower harmonics. But it was simply impossible to measure pulses of higher harmonics around 100 eV photon energy with this technique. Later, cross correlation techniques were employed. Attosecond XUV pulses were used to photoionise gases and the energy of photoelectrons was measured. The spectrum of the photoelectrons mimics that of the • • •

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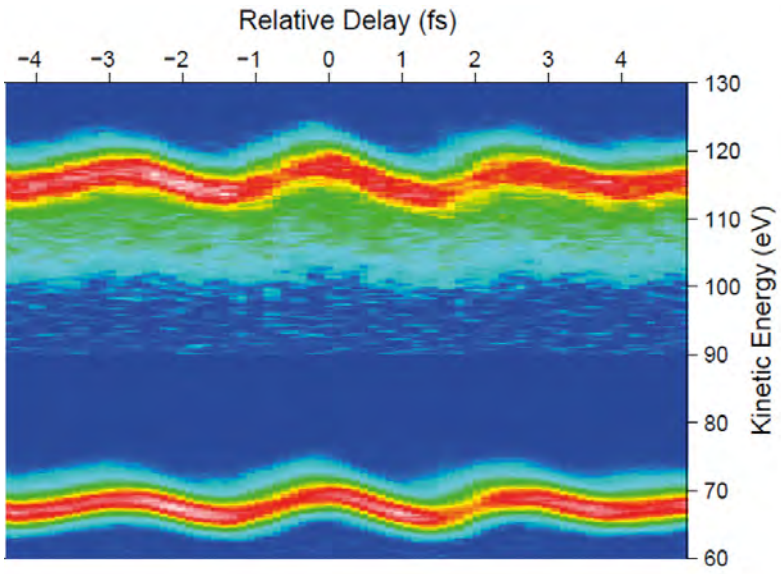
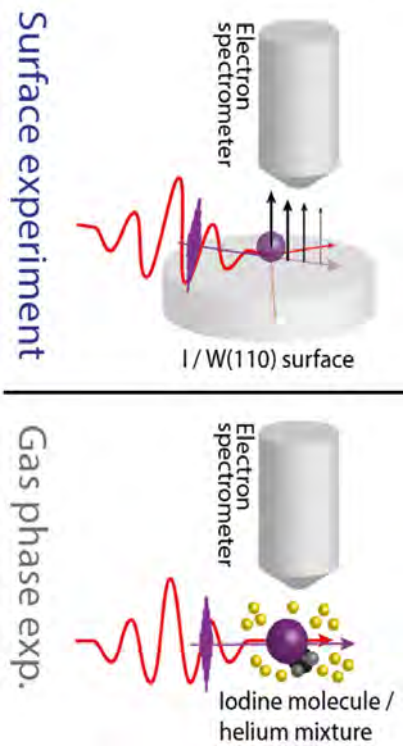


▲ **FIG. 3:** Streaking spectrograms of two different photoelectrons, *e.g.* conduction band and core level electrons, ending up at different kinetic energies due to different ionization potentials. The temporal shift between the two spectrograms corresponds to the relative delay in photoemission.

harmonics, just reduced by the ionization potential of the atom. A dressing IR field modulates the spectrum of the photoelectrons. When attosecond XUV pulse trains are used for photoionization and multi-cycle IR pulses as the dressing field, one obtains sidebands in the photoelectron spectrum. Using the HHG driver pulses as the dressing field, these sidebands are situated exactly between the harmonics. Harmonic N plus one IR photon gives the same energy as harmonic N+2 minus one IR photon (*nota bene*, in gases only odd harmonic orders are generated). This creates an interference which – by changing the delay between the attosecond pulse train and the dressing pulses – enables to measure the duration of the individual XUV pulses. With this method called "Reconstruction of attosecond beating by interference of two-photon transitions" (RABBITT) Pierre Agostini and his group were able to measure the first attosecond pulse trains in 2001, where the individual pulses in the train were as short as 250 asec [6]. RABBITT can not only be used to characterize attosecond pulse trains but since then it was employed to measure electronic processes on the attosecond time scale.

A very special feature in HHG appears when the driving laser pulse comprises only few cycles of the electric field. Nowadays pulses with just a little bit more than one optical cycle are used and the phase between the electric field and the intensity envelope of the pulse ("carrier envelope phase") is set to an optimum value. The latter was made possible by the work of Theodor Hänsch and John Hall that yielded their Nobel Prize in 2005. The shortness of few-cycle pulses implies that the photoemission / recombination-process as described above happens just a few times and that the harmonics generated "at the center" of the pulses end up with higher energies compared to the harmonics generated at neighbouring field oscillation cycles. If one selects the energetically highest harmonic and discards all the rest of the XUV radiation, one gets a single, isolated attosecond pulse. The measurement of these pulses uses a similar method as RABBITT, namely a cross correlation between the XUV pulse and a dressing electric field in photoelectron spectroscopy. Since there is only one attosecond XUV pulse, which is shorter than half the oscillation cycle of the dressing IR pulse, there is no interference of multiple pulses any more. Photoelectrons generated by the attosecond XUV pulse just "feel" the electric field of the dressing IR pulse at the actual delay. The photoelectrons are energetically up- and downshifted ("streaked") following the electric field – to be correct: the vector pontential – of the dressing pulse and generate a spectrogram for varying delay. With this method, "attosecond streaking spectroscopy", the group of Ferenc Krausz succeeded to measure the first single isolated attosecond pulses, having a duration of 650 as, in 2001 [7]. Attosecond streaking spectroscopy, too, is used to measure electron dynamics in gases, molecules, solids and layered systems, since then [8].

A seminal experiment on tungsten [9] where a delay in photoemission of conduction band electrons and core level electrons was observed triggered many theoretical activities leading to different explanations on the physical reason of the delay. From [10] we learned that a dealy in photoemission even happens in atoms, here the 2s and 2p electrons in Neon. Attosecond streaking experiments have been performed on different solids, layered structures and liquids, resulting in different delays – also depending on the excitation photon energy. These measurements lead to a stepwise increase of the understanding of different physical effects contributing to the timing of photoemission. Also, these experiments shed light on fundamental processes like the dielectric screening of electric fields in a solid. Since attosecond streaking measurements always



◀ **FIG. 4:** Measurement of the attosecond photoemission delay from tungsten and calibrating it by referencing the delay to iodine and helium delay, respectively.

yield relative delays of photoemission events (Figure 3), it was very important to calibrate the method to Helium which could fully be calculated. This way, the measurement of absolute emission delays were made possible (Figure 4).

Attosecond science thus evolved from the basic concepts of nonlinear optics and intriguing experimental observations via the development of ultrashort XUV pulse measurement techniques reaching attoscience applications more recently. As a result, the measurement of electron dynamics at their natural attosecond timescale is now available in many laboratories around the world, "for the greatest benefit to humankind". ■

Acknowledgements

The authors acknowledge Ferenc Krausz for his guidance and mentorship during their early career. PD acknowledges the National Office for Research, Development and Innovation of Hungary for support (projects 137373 and TKP2021-NVA-04).

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References

[1] P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich, *Phys. Rev. Lett.* **7**, 118 (1961).

[2] A. L'Huillier, Ph. Balcou, *Phys. Rev. Lett.* **70**, 774 (1993)

[3] J.L Krause, K.J. Schafer and K.C Kulander, *Phys. Rev. Lett.* **68**, 3535 (1992).

[4] P. B. Corkum, *Phys. Rev. Lett.* **71**, 1994 (1993)

[5] G. Farkas and C. Tóth, *Phys. Lett. A* **168**, 447 (1992).

[6] P.M. Paul *et al.* Science **292**, 1689 (2001).

[7] M. Hentschel, R. Kienberger *et al.*, *Nature* **414**, 509 (2001).

[8] R. Kienberger *et al.*, *Nature* **427**, 817 (2004)

[9] A. Cavalieri *et al.*, *Nature* **449**, 1029 (2009)

[10] M. Schultze *et al.*, *Science* **328**, 1658 (2010).