Journal of Physics B: Atomic, Molecular and Optical Physics

VIEWPOINT

Multiple harmonic conversion of 1064 nm in rare gases

To cite this article: Anne L'Huillier 2017 *J. Phys. B: At. Mol. Opt. Phys.* **50** 060501

IOP Publishing

Journal of Physics B: Atomic, Molecular and Optical Physics

J. Phys. B: At. Mol. Opt. Phys. **50** (2017) 060501 (3pp)

https://doi.org/10.1088/1361-6455/aa5935

Viewpoint

Multiple harmonic conversion of 1064 nm in rare gases

Anne L'Huillier
Department of Physics, Lund University, Sweden

This viewpoint relates to an article by M Ferray *et al* (1988 *J. Phys. B: At. Mol. Opt. Phys.* **21** L31–L35) and is published as part of a series of viewpoints celebrating 50 of the most influential papers published in the *Journal of Physics* series, which is celebrating its 50th anniversary.

Background and motivation

Harmonic generation in gases was studied extensively in the late 1970s with the aim of reaching short wavelengths as well as high conversion efficiencies. Most of this research used ultraviolet lasers to reach vacuum ultraviolet radiation through low-order processes such as third- or fifth-harmonic generation. In parallel to these developments, a few groups worldwide studied multiphoton ionization in strong laser fields. Interesting phenomena such as multiple ionization or above-threshold ionization were observed as high-power laser technology progressed, thereby enabling high intensities to be reached, moving the light−atom interaction regime beyond the weak field (perturbative) regime. A natural extension of these experiments was to look at the fluorescence spectra emitted when an atomic gas is exposed to a strong laser field in order to get information about the excitation processes in the medium. This was the motivation for the experiments discussed in this viewpoint [1] (see also related experiments reported in [2]). The experiments did not show what was expected; instead, a new phenomenon, high-order harmonic generation (HHG), was discovered.

Experimental discovery

The experiments used a commercial Nd:YAG mode-locked laser with pulse duration of 30 ps, repetition rate 10 Hz, wavelength 1064 nm in the infrared (IR) region, 30 mJ energy, allowing us to reach focused intensities of a few times 10^{13} W cm^{-2}. The gas medium was provided by a gas jet and the detection system used a monochromator imaging the gas jet and an electron multiplier. In the experiment, a grating was rotated to scan the emission wavelength–in contrast to today's experimental setups where a spectrum can be recorded at once using a flat-field spectrometer. The observed spectra in Xe, Kr and Ar consisted of a series of odd-order harmonics of the laser field up to a very high order (21 in Xe, 29 in Kr, 33 in Ar). Due to the long pulses used to drive the generation process, the harmonic spectral width was quite narrow. The harmonics were found to be emitted along the laser propagation axis with a small divergence angle.

The main result of the article was the observation of the characteristic behavior of these high harmonics: first a decrease for the first orders, then a plateau where the harmonics have similar yields and next a rather sharp atom-dependent cutoff. This result directly led to the question: why are very high-order harmonics generated and why does the harmonic strength stay almost constant in spite of such different nonlinear orders? The article did not explain the origin of the high-order harmonics but concluded that high-order harmonic generation (HHG) happens in similar conditions as multiphoton ionization and so must be related to it. The unexpected effect of fundamental laser wavelength on the generation of very short wavelength radiation was also pointed out.

Prediction and interpretation

Almost directly after the publication of the article, it was suggested that if all these harmonics were emitted 'in phase' their interference would lead to the formation of very short pulses, in the attosecond range (1 as $= 10^{-18}$ s), in analogy to the mode-locking technique in lasers [3, 4]. In parallel to the attosecond question, numerical methods were developed to solve the time-dependent Schrödinger equation for an atom in a strong laser field [5] as well as the propagation equations [6] in order to gain some insight into the physics of HHG. An empirical law was proposed for the position of the cutoff [7], which triggered a beautiful semi-classical interpretation of HHG [8, 9]. In this picture, the laser field induces a distortion of the atomic potential so that an electron can be ionized by the tunnel effect and driven away from the core by the laser field. When the field changes sign, the electron is attracted towards the ionic core. It may return with a high kinetic energy and be captured by its parent ion, emitting an XUV photon. Everything happens within a laser optical cycle and the emitted light pulse has a width of a few hundred attoseconds. This coherent phenomenon follows the oscillations of the laser field and is repeated at each optical half cycle. Interference between consecutive attosecond pulses results in a frequency comb of high-order (odd) harmonics. The semi-classical model could explain the cutoff, the high-order harmonic plateau, the relation between strong-field (tunnel) ionization and HHG, and predict the formation of attosecond pulses.

HHG today: sources and applications

HHG became, and is still, an active field of research worldwide. Over many years, amplified femtosecond titanium sapphire lasers, which became available just a few years after the experiments reported in [1, 2], were the 'standard' laser for HHG. Today there is an increased diversity of HHG sources driven by a variety of lasers ranging from high energy lasers at low repetition rates to high average power lasers, based upon optical parametric amplification or simply high-power oscillators. HHG sources can be vastly different [10] and should be designed for specific applications. There is also a lot of effort being devoted to the extension of HHG sources to higher photon energies in the soft x-ray range, in particular using mid-IR sources [11, 12].

HHG remained for a long time 'a solution looking for a problem', in terms of applications. The relatively low efficiency of the process made applications demanding two-photon absorption (e.g, XUV pump/XUV probe experiments) difficult [13, 14]. There is now, however, increasing activity in this direction. In spite of the early prediction, the experimental demonstration of the existence of attosecond pulses had to wait 14 years [15, 16]. Since then, the field of attosecond science utilizing either single attosecond pulses, attosecond pulse trains or even the HHG process itself, is expanding worldwide [17]. In general, we see an increased diversity of HHG applications ranging from fundamental attosecond science to coherent diffraction imaging of lithographic or biological samples [18], spanning many areas from atomic and molecular physics to condensed matter.

Personal remarks

From a more personal point of view, this work was determinant for my research career. I was at the time a young researcher, who had just gotten a permanent researcher position but did not have a clear research direction. I became fascinated by this phenomenon, at the border of atomic physics and nonlinear optics, including both a microscopic fundamental aspect: the response of an atom to a strong laser field and a more applied aspect: the buildup of the harmonic field in the nonlinear medium. There was also the exciting perspective of supporting the shortest light pulses ever produced. Today, almost 30 years later, I am still working on HHG and its applications. I feel highly privileged to have had the chance to participate in this research from the beginning.

The author acknowledges support from the European Research Council (Advanced grant PALP) and the Swedish Research Council.

References

[1] Ferray M *et al* 1988 *J. Phys.* B **21** 31
[2] McPherson A *et al* 1987 *J. Opt. Soc. Am.* B **4** 595
[3] Farkas G and Toth C 1992 *Phys. Lett.* A **168** 447
[4] Harris S E, Macklin J J and Hänsch T W 1993 *Opt. Commun.* **100** 487
[5] Kulander K C and Shore B W 1989 *Phys. Rev. Lett.* **62** 524
[6] L'Huillier A *et al* 1992 *Phys. Rev.* A **46** 2778
[7] Krause J L, Schafer K J and Kulander K C 1992 *Phys. Rev. Lett.* **68** 3535
[8] Schafer K J *et al* 1993 *Phys. Rev. Lett.* **70** 1599
[9] Corkum P B 1993 *Phys. Rev. Lett.* **71** 1994
[10] Heyl C M, Arnold C L, Couairon A and L'Huillier A 2017 *J. Phys.* B **50** 013001
[11] Colosimov P *et al* 2008 *Nat. Phys.* **4** 386
[12] Popmintchev T *et al* 2012 *Science* **336** 1287
[13] Tzallas P *et al* 2003 *Nature* **426** 267
[14] Nabekawa Y *et al* 2005 *Phys. Rev. Lett.* **94** 043001
[15] Paul P M *et al* 2001 *Science* **292** 1689
[16] Hentschel M *et al* 2001 *Nature* **414** 509
[17] Krausz F and Ivanov M 2009 *Rev. Mod. Phys.* **81** 163
[18] Zhang B *et al* 2015 *Ultramicroscopy* **158** 98