Appl. Phys. B 78, 845–849 (2004)
DOI: 10.1007/s00340-004-1478-6

Applied Physics B
Lasers and Optics

C. VALENTIN✉
S. KAZAMIAS*
D. DOUILLET
G. GRILLON
T. LEFROU
F. AUGÉ
S. SEBBAN
P. BALCOU

Experimental observation of anomalous high harmonics at low intensities

Laboratoire d'Optique Appliquée, ENSTA – Ecole Polytechnique, CNRS UMR 7639, Chemin de la Hunière, 91761 Palaiseau, France

Received: 31 October 2003/Revised version: 19 January 2004
Published online: 30 March 2004 •

ABSTRACT A variety of complex phenomena occurs when an ultra-short laser pulse interacts with atoms in the 10^{13} W/cm^2 range: non-sequential ionisation, electron recollision, and Freeman resonances. We show that high-harmonic spectra obtained experimentally in this parameter range also display anomalous features, which are difficult to understand in the framework of the three-step, semiclassical model. The results of a systematic study of these high harmonics generated in argon, xenon, and krypton are presented. From the experimental curves, complex high-order harmonic generation phenomena are discussed.

PACS 42.65.Ky; 32.80.Rm

1 Introduction

High-harmonic generation (HHG) occurs when an ultra-intense and ultra-short laser beam interacts with a gaseous medium. In this case, only odd harmonics are generated. Their spectra present a characteristic shape: the efficiency of the first perturbative orders decreases rapidly; then, their intensity is roughly equal along the plateau region, which is finished by a sudden cutoff. This spectral shape is theoretically understood thanks to the three-step model [2, 3]. When the laser electric field is high enough to lower the atomic Coulomb barrier, one outer electron is freed by tunnel ionisation. The resulting free electron is then driven by the laser electric field; as the latter changes sign, the electron may come back to the vicinity of the core. When it recombines radiatively with its parent ion, one harmonic photon is emitted. From this model, a general cutoff law has been derived: $N_{\max}\hbar\omega = I_p + 3.2U_p$, where $N_{\max}$ is the maximum order that can be reached, I_p is the ionisation potential of the atom considered, and U_p is the ponderomotive energy which depends linearly on the laser intensity. The factor 3.2 corresponds to the maximum kinetic energy that the electron can acquire in the continuum. The theoretical maximum order is then given by the cutoff law applied with the barrier suppression intensity (I_{BSI}), which is the intensity at which the laser electric field compensates the atomic Coulomb barrier. This simple model has been quantically generalised by Lewenstein et al. [4]. Over ten years, the theoretical predictions have been experimentally validated [5]. Nevertheless, there were still a few experiments where anomalous high orders were reported [6–8]. These high orders have been explained in terms of non-adiabatic ionisation or by harmonics generated by ions created by sequential ionisation.

The aim of this article is to present experimental data obtained with a very sensitive spectrometer for short wavelengths, demonstrating anomalously high harmonic spectra. They have been observed in very specific experimental conditions, quite different from those optimising the harmonic source [9]. We first describe the experimental setup, and especially the spectrometer and its performance. Then, we show the experimental spectra we have recorded thanks to this spectrometer. A systematic study of harmonic signal versus position of the gaseous medium with respect to the focus position, pressure in the cell, and infrared laser energy is presented. Finally, we propose open suggestions in order to explain these surprising data.

2 Experiment and results

2.1 *Experimental setup*

The experimental setup for generating high harmonics is described in [9]. The LOA 'Salle Rouge' system is a titanium : sapphire laser which delivers pulses at 805 nm with 30-fs duration and 6-mJ energy at a repetition rate of 1 kHz. The energy of the fundamental laser can be modified by turning a half-wave plate placed before a polariser in the laser chain. The infrared beam can be apertured by an iris, before it is focused by a MgF_2 lens with 1-m focal length. When the iris is fully opened, the Rayleigh range is rather small ($\approx$ 2.5 mm) and the whole energy is transmitted. Closing the iris down to 14 mm results in an increase of the Rayleigh range ($\approx$ 11 mm); the transmitted energy is only 55% [9, 10]. The focused laser beam interacts with a gaseous cell filled with argon, xenon, or krypton where HHG takes place. The cell length can be varied, as can the position of its entrance with respect to the focus. The pressure in the cell can also be adjusted from 0 to 50 Torr. Then the harmonic beam propagates collinearly with the infrared beam. In order to avoid any infrared radiation in the detection system, we insert two

✉ Fax: +33-1/6931-9996, E-mail: valentin@ensta.fr
Present address: Laboratoire d'Interaction X Avec la Matière, Bât. 350, Université Paris-Sud, 91405 Orsay, France

aluminium filters with 250-nm thickness. They cut totally the IR beam and transmit XUV radiation from 17 nm to 80 nm corresponding to orders from the 47th to the 11th. The experimental value of the one-filter transmission is 44% at 18 nm (H45), decreasing to 22% at 32 nm (H25). The harmonic radiations are then analysed by a spectrometer composed of a toroidal gold mirror and a 600-gr/mm grating at grazing incidence. The mirror focuses in both directions to enhance the sensitivity of the detection system. The theoretical reflective factor for the mirror is about 60% for a large spectral range. The diffracting efficiency for the first order of the grating calculated by the manufacturer is 10% at 10 nm and smoothly decreases to 4% at 50 nm. Considering both the spectrometer and the filter transmission, the measuring sensitivity is 10 times higher at 18 nm (H45) than at 32 nm (H25). The spectra are recorded thanks to a cooled XUV CCD camera with an integration time between 0.1 and 10 s, corresponding to 100 to 10^4 shots for the 1-kHz laser repetition rate. The measured spectra are obtained by merging two (for Ar) or three (for Xe and Kr) spectra measured at different camera angles with respect to the grating. Considering the signal to noise ratio equal to 1 for the camera (four shots per 50 pixels), the minimum detectable signal can reach 3×10^{-2} photons per shot per high harmonic at the source.

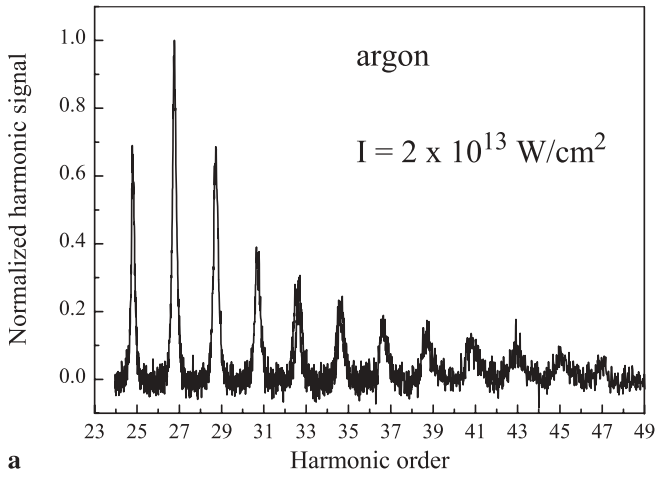
This reflection spectrometer is therefore very sensitive to short wavelengths, allowing us to detect extremely faint harmonic signals.

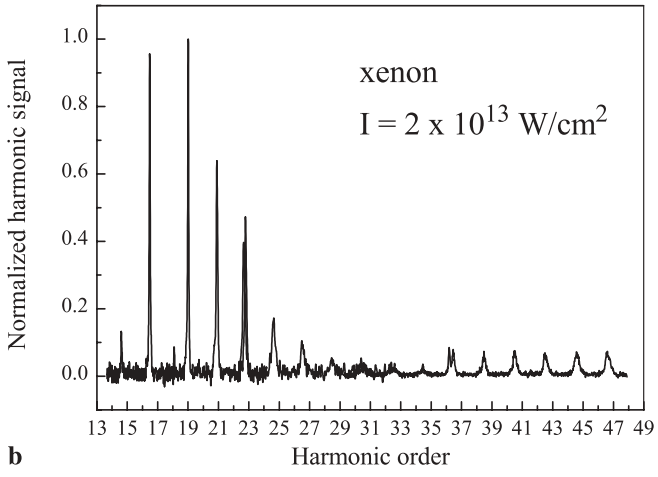
2.2 *Spectra description*

We display in Fig. 1 three spectra obtained for argon, xenon, and krypton when the cell is placed as much as 25–30 mm after the focus position, and the iris is fully open. The fundamental laser energy was varied between 3.6 and 4.2 mJ. For a non-apertured laser beam (Rayleigh range $z_{\text{Rayleigh}} = 2.5$ mm), the corresponding laser intensity in the cell is approximately 2×10^{13} W/cm^2. The relative intensity uncertainty is mainly due to the uncertainty Δz_{cell}: for $\Delta z_{\text{cell}} = 5$ mm, the calculated relative uncertainty is 33%. The shot to shot fluctuations of the laser energy are not taken into account because the measured energy is given for a long integration time (typically 10 s). For apertured beams, the beam profiles for different positions with respect to the focus have been computed by considering the phase measured at focus using a Shack–Hartmann sensor [9]. In this case, the over-intensities would lead to intensity uncertainties of up to a factor of two. The spectra are spectrally calibrated considering the aluminium filter cutoff at 17 nm.

The spectrum displayed in Fig. 1a is obtained with 25 Torr of argon in a 2-mm-length cell. The maximum signal is recorded for the 27th order (≈ 42 eV); it then drops, before it decreases smoothly until the 47th one, corresponding to the aluminium filter transmission cutoff at 17 nm. For argon, the reabsorption of the low harmonics is very high at the gas density used in the cell, so we have not detected harmonics lower than the 23th order.

However, the usual spectra for argon in the absorption limit present a completely different shape, with a relatively small number of harmonic orders (H23 to H29) dominating completely the spectrum [10]. This behaviour is usually attributed to the Cooper minimum in the absorption cross section of argon around those wavelengths. One may note that such high orders have already been detected in argon in other experiments [7], but with a markedly different and smoother shape, and laser intensities in the medium over one decade su-





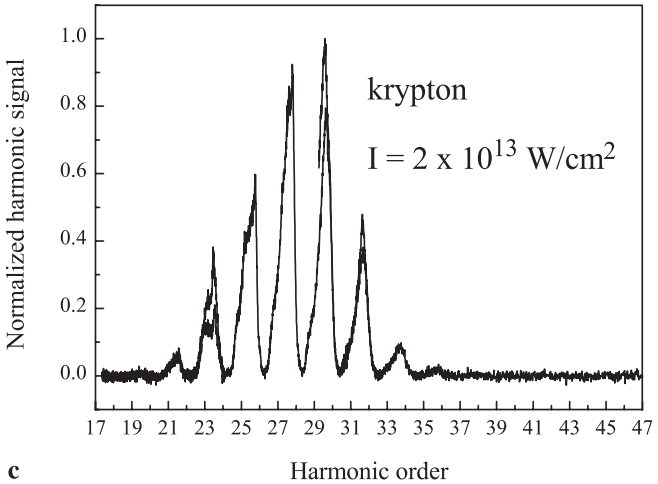


FIGURE 1 All the spectra are obtained with ≈ 4-mJ laser energy and an iris diameter of 40 mm ($z_{\text{Rayleigh}} = 2.5$ mm); the cell entrance is placed at ≈ 25 mm from the laser focus leading to an intensity of $\approx 2 \times 10^{13}$ W/cm^2. **a** Spectrum obtained with 2-mm cell filled with 25 Torr of argon, **b** spectrum obtained with 3-mm cell filled with 10 Torr of xenon, **c** spectrum obtained with 2-mm cell filled with 15 Torr of krypton

perior to those used here. The explanation proposed was the existence of non-adiabatic ionisation effects.

More surprising is the spectrum obtained for 10 Torr of xenon in a 3-mm-length cell (Fig. 1b). It is more difficult to reach such high harmonics with xenon atoms because orders beyond the 17th cannot theoretically be reached (cutoff law with I_{BSI}). The maximum signal is measured for the 19th order (≈ 30 eV) and, as for the argon spectrum, the signal decreases until the 35th. Then, it presents a second plateau between H37 and at least H47, this limitation being due to the aluminium filters. The experimental intensity in the cell is clearly too low to reach such high orders considering the cutoff law from the three-step model. Similarly to the argon spectrum, the lower orders are reabsorbed by the gas. Moreover, calculating the 10 Torr xenon gas transmission for a 3-mm-length cell, we find that the spectrum is not observed in the absorption limit conditions for high orders [11].

Finally, for krypton (cf. Fig. 1c), the spectrum presents a totally different shape: only orders from the 21th to the 33th are observed. Above 40 nm (H21), the harmonic radiations are probably reabsorbed by the gas itself. The highest reached order is above the I_{BSI} cutoff order for krypton.

It must be stressed than the maximum number of photons is about 4×10^2 photons per shot for H39 in argon, which is five orders of magnitude lower than the H23 signal generated in argon with the usual conditions of generation. Whatever physical process explains these harmonic spectra is therefore quite inefficient. In the specific case of xenon, we had indeed to resort to exposure times up to 10 s to get the signal.

2.3 *Systematic study*

In order to explain these surprising spectra, we have performed a systematic study. First, in Fig. 2, we show the curve for the H33 and H45 signals versus the position of the cell entrance with respect to the focus position for 15 Torr of argon. For the highest intensity at the focus (2.8×10^{14} W/cm^2 in vacuum), H33 belongs to the plateau, but not H45. Experimentally, standard spectra are obtained when the laser is focused in the medium, where the intensity is maximum, or when the cell is placed before the focus; in particular, H45 is not detected. In contrast, the H45 signal is a a maximum when the cell is placed 25 mm after the focus, in which place the intensity is only 6.8×10^{13} W/cm^2. However, the laser intensity in the medium should increase when the focus gets closer to the cell, whereas the H45 signal decreases while the H33 signal increases. This behaviour cannot be explained by the cutoff law.

Second, we have performed a study of the H39 signal versus pressure in the cell for argon (Fig. 3). The signal linearly increases until 25 Torr and then it saturates. This curve shows that the harmonics are still generated by atoms or ions, but not by dimers or clusters [12, 13]. In this case, we would have expected a power law instead of a linear rising of the signal.

Third, we show in Figs. 4 and 5 harmonic signals versus the fundamental laser energy for argon and xenon. For argon, the three harmonic signals (H29, H33, and H45) increase linearly with the energy; then they saturate and even the H33 and H45 ones decrease when the intensity reaches the 10^{14} W/cm^2 range. For xenon, the H21 and H45 signals increase non-linearly. The H21 signal follows a power law of 0.5 and the H45 one follows a power law of 3. For all the curve points, the experimental intensity is well below the cutoff intensity. From Lewenstein's theory, we expect an exponential law when the harmonics belong to the cutoff region. It seems that the highest harmonics do not follow the cutoff law, considering the asymmetric curve in Fig. 3 and the power-law rise of the harmonic signal belonging the cutoff region. We can conclude that this theory is not suitable to predict such a behaviour. Different behaviours for argon and xenon atoms should be due to different atomic structures. From these experimental results, it is important to underline the complexity of the generating process.

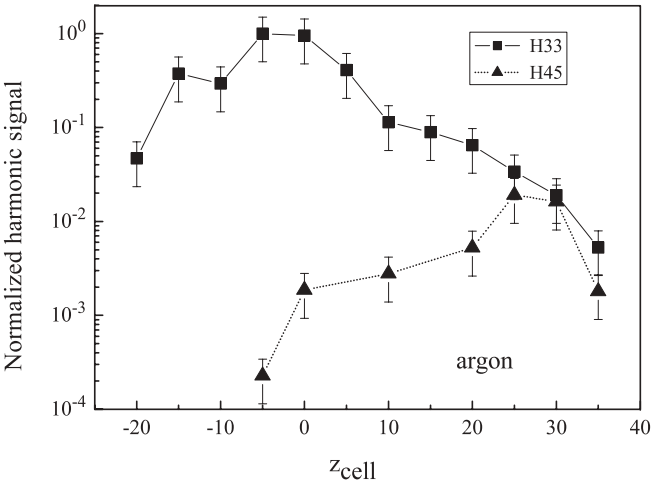


FIGURE 2 Signals of 33th and 45th harmonic orders generated in 2-mm cell filled with argon versus the focus position with respect to the cell entrance. The laser energy is 3.45 mJ, the aperture diameter is 14 mm ($z_{Rayleigh} = 11$ mm), and the pressure is 15 Torr. The intensity in the cell varies from 0.3×10^{14} W/cm^2 (at $z_{cell} = 35$ mm) to 2.8×10^{14} W/cm^2 (at $z_{cell} = 0$)

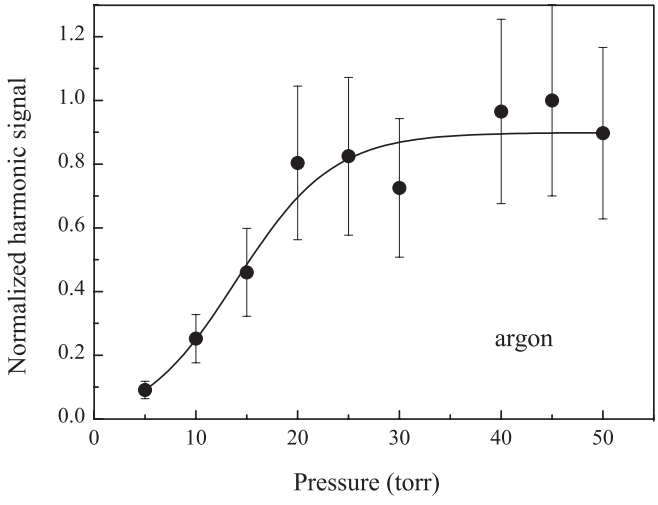


FIGURE 3 Signal of 39th harmonic order generated in 2-mm cell filled with argon versus pressure. The laser energy is 3.65 mJ, the aperture diameter is 14 mm, and the cell entrance is located 25 mm after the focus leading to an intensity of 5×10^{13} W/cm^2 in the medium

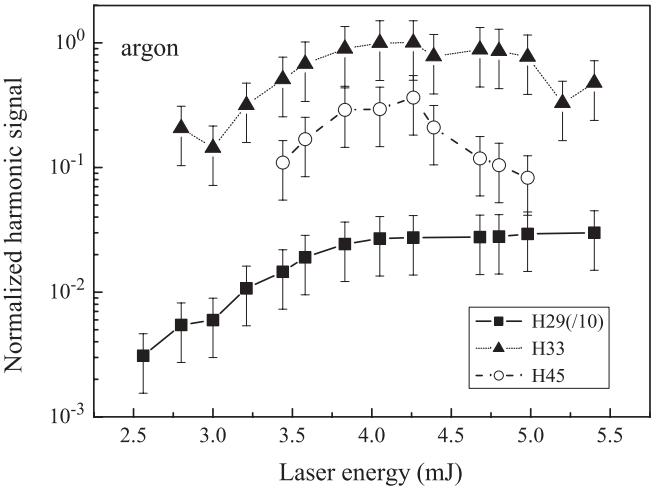


FIGURE 4 Signals of 29th divided by ten, 33th, and 45th harmonic orders generated in 2-mm cell filled with 15 Torr of argon versus fundamental laser energy. The aperture diameter is 14 mm and the cell entrance is located 22 mm after the focus position. The intensity in the medium is 7×10^{13} W/cm^2 for a laser energy of 4 mJ

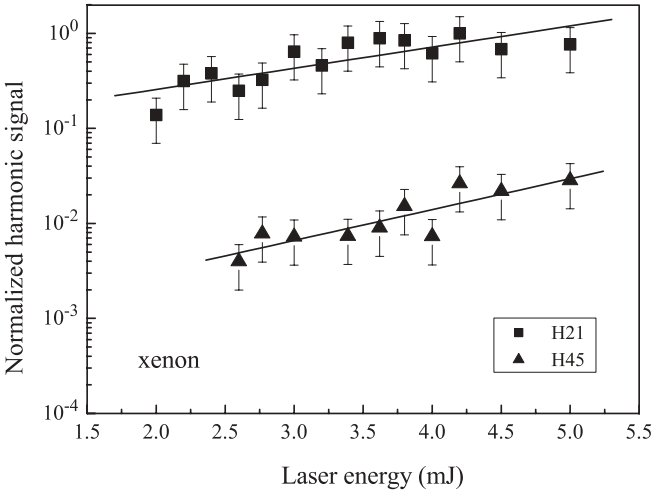


FIGURE 5 Signals of 21th and 45th harmonic orders generated in 3-mm cell filled with 10 Torr of xenon versus fundamental laser energy. The aperture diameter is 40 mm and the cell entrance is located 20 mm after the focus position. The intensity in the medium is 5×10^{13} W/cm^2 for a laser energy of 5 mJ

3 Discussion

A few possibilities can be discussed to explain these experimental results. The first is to invoke harmonic generation by ions. Such a process has indeed been seen recently, using modulated hollow-core fibers [14]. In our setup, it might be possible to generate harmonics by ions at the laser focus where the intensity in vacuum should be in the 10^{15} W/cm^2 range, which means several centimetres out of the gas cell. The gas density in this region is very low (estimated to be in the 10^{-2} mbar range), and harmonic generation depends on the square of the density, making this process extremely unlikely. Considering Fig. 2, the drop of the H45 signal when the laser focus is located at the cell entrance could then be the result of the laser-beam defocusing. Some experimental features are not fully consistent with this ion-emission scenario. First, in Fig. 2, beam defocusing is not expected at all when the focus is located as much as 10 or 20 mm before the cell; the signal level of H45 in those two positions is however already strongly reduced with respect to the maximum at $z = 25$ mm. Second, one expect the intensity dependences of H45 generated by ions, and H21 generated by atoms, to be very strongly different. Figure 5 shows that this is not the case.

We investigate another hypothesis to explain these anomalous high-order harmonics. We notice that, at an intensity of 2×10^{13} W/cm^2, the ponderomotive energy U_p is only 1.2 eV, which is very small compared to the ionisation potential I_p for the three atomic species. Considering the Keldysh parameter [15], which is over 1 in this case, the ionisation process is a complex mixing of tunnelling ionisation and IR multiphoton ionisation.

In order to increase the maximum order, one can consider the three terms of the cutoff law: increasing the factor k before the ponderomotive energy U_p, increasing the ponderomotive energy, and/or increasing the ionisation potential. To increase the factor k, it could be possible to consider a recollision of the electron with its parent ion [16]. A calculation inspired from Lewenstein's theory leads to a factor equal to 5.5 instead of 3.2. The recollision effect on the maximum order (N_{max}) is not enough to explain very high orders. To increase U_p, one should increase the laser intensity or increase its wavelength. In our case, the wavelength is fixed and very high harmonics have been observed only for very low intensities ($< 10^{14}$ W/cm^2 range). Consequently, the increase of the ionisation potential would lead to higher N_{max}. Non-sequential multi-ionisation [17], a shake-off effect [18], or photo-ionisation by absorption of lower harmonics are the processes that could be involved to increase I_p.

For the Kr spectrum, it appears that the harmonics could generated by two charged krypton ions, because the 25th order corresponds to the energy of Kr^{2+} ($I_p = 37$ eV [19]) with $U_p = 1.2$ eV. Such two charged ions could result from the process of non-sequential ionisation, shown to exist in this parameter range. For the Ar and Xe spectra, a more complex process could involve ions in a hollow core state. The energy of this state corresponds to the energy of the maximum signal order (e.g. the 27th for Ar equal to 42 eV). This state should be populated by a shake-off process with half the laser periodicity and depopulated by radiative recombination towards the fundamental ionic state faster than Auger radiative decay [20].

4 Conclusion

We have observed and studied anomalous high-harmonic orders generated in argon, xenon, and krypton when the laser focus is located far from the gaseous medium. The spectral shapes are completely different than those observed in the usual configuration of laser intensity and interaction geometry, depending on the studied noble gas. The measured signals for high orders lead to very low conversion efficiencies (a few 10^{-12}). These high orders can then be detected when using a very sensitive spectrometer coupled to a cooled XUV CCD camera, with long exposure times.

Our data may be the first to show the need to go further than the universally accepted, and extremely effective Lewenstein model, in specific conditions. One could invoke more

complex processes for HHG: non-sequential multi-ionisation, a shake-off process, or electron collision and rescattering before the radiative recombination leading to harmonic generation. Such possibilities should be theoretically addressed, keeping in mind that there is no reason why the different behaviours observed in the three heavy noble gases (Xe, Kr, and Ar) should all have exactly the same interpretation. This limits any prospect of applying this anomalous HHG radiation in the short term.

ACKNOWLEDGEMENTS We gratefully acknowledge Christian Brach from Jobin-Yvon for the diffraction-efficiency computation of the grating and Philippe Zeitoun from LIXAM (Laboratoire d'Interaction X Avec la Matière) and LOA (Laboratoire d'Optique Appliquée) for the loan of the XUV CCD camera. Some theoretical models hinted at here were elaborated in collaboration with M. Lewenstein and J.-F. Wyart, and will be detailed elsewhere.

REFERENCES

1 L.F. DiMauro, P. Agostini: 'Ionization Dynamics in Strong Laser Field', In: *Advances in Atomic, Molecular and Optical Physics, Vol. 35*, ed. by B. Bederson, Walther (Academic, London 1995) p. 79
2 K.C. Kulander, K.J. Shafer, J.L. Krause: 'Dynamics of short-pulse excitation, ionization and harmonic conversion', In: *Super-Intense Laser–Atom Physics* (NATO ASI B316), ed. by P. Piraux, A. L'Huillier, K. Rzążewski (Plenum, New York 1993) p. 95
3 P.B. Corkum: Phys. Rev. Lett. **71**, 1994 (1993)
4 M. Lewenstein, P. Balcou, M.Yu. Ivanov, A. L'Huillier, P.B. Corkum: Phys. Rev. A **49**, 2117 (1994)
5 A. L'Huillier, M. Lewenstein, P. Salières, P. Balcou, M. Ivanov, J. Larsson, C.-G. Wahlström: Phys. Rev. A **48**, R3433 (1993)
6 C.-G. Wahlström, J. Larsson, A. Persson, T. Starczewski, S. Svanberg, P. Salières, P. Balcou, A. L'Huillier: Phys. Rev. A **48**, 4709 (1993)
7 I.P. Christov, J. Zhou, J. Peatross, A. Rundquist, M.M. Murnane, H.C. Kapteyn: Phys. Rev. Lett. **77**, 1743 (1996)
8 N.A. Papadogiannis, C. Kalpouzos, E. Gouliemakis, G. Nersysian, D. Charalambidis, F. Augé, F. Weihe, P. Balcou: Appl. Phys. B **73**, 687 (2001)
9 S. Kazamias, F. Weihe, D. Douillet, C. Valentin, T. Planchon, S. Sebban, G. Grillon, F. Augé, D. Hulin, Ph. Balcou: Eur. Phys. J. D **21**, 353 (2002)
10 S. Kazamias, D. Douillet, C. Valentin, Th. Lefrou, G. Grillon, G. Mullot, F. Augé, P. Mercère, Ph. Zeitoun, Ph. Balcou: Eur. Phys. J. D **26**, 47 (2003)
11 http://www-cxro.lbl.gov
12 C. Lyngå, A. L'Huillier, C.-G. Wahlström: J. Phys. B **29**, 3293 (1996)
13 T.D. Donnelly, T. Ditmire, K. Neuman, M.D. Perry, R.W. Falcone: Phys. Rev. Lett. **76**, 2472 (1996) and references therein
14 E.A. Gibson, A. Paul, N. Wagner, R. Tobey, S. Backus, I. Christov, M.M. Murnane, H.C. Kapteyn: Phys. Rev. Lett **92**, 033001 (2004)
15 L.V. Keldysh: Sov. Phys. JETP Lett. **1**, 66 (1965)
16 R. Kopold, W. Becker, H. Rottke, W. Sandner: Phys. Rev. Lett. **85**, 3781 (2000)
17 S. Larochelle, A. Talebpour, S.L. Chin: J. Phys. B **31**, 1201 (1998)
18 D.N. Fittinghoff, P.R. Bolton, B. Chang, K.C. Kulander: Phys. Rev. Lett. **69**, 2642 (1992)
19 R.D. Cowan: 'Experimental background and basic concepts', In: *The Theory of Atomic Structure and Spectra*, ed. by D.H. Sharp, L.M. Simmons Jr. (University of California Press, Berkeley 1981) p. 12
20 M. Drescher, M. Hentschel, R. Kienberger, M. Uiberacker, V. Yakovlev, A. Scrinzi, Th. Westerwalbesloh, U. Kleineberg, U. Heinzmann, F. Krausz: Nature **419**, 803 (2002)