Physics Letters A 168 (1992) 447–450
North-Holland

PHYSICS LETTERS A

Proposal for attosecond light pulse generation using laser induced multiple-harmonic conversion processes in rare gases

Gy. Farkas and Cs. Tóth

Research Institute for Solid State Physics, Central Research Institute for Physics, P.O. Box 49, H-1525 Budapest, Hungary

Received 11 June 1992; accepted for publication 13 July 1992
Communicated by V.M. Agranovich

A new principle of attosecond light pulse generation is suggested. The method is based on a Fourier synthesis of laser induced multiple harmonics, which all are oscillating with the same fixed phase as predicted and observed recently in rare gases. According to our calculation using published experimental data, the production of a regular sequence of ~30–70 as duration light pulses is expected to be realizable.

Recent studies on ultrashort-scale light–matter interactions have stimulated the elaboration of new procedures for producing extremely short light pulses.

The most effective of these procedures is based on the Fourier synthesis of the equidistant components of a given spectrum range $\Delta\omega$ of a light emitting source. Therefore the shortest achievable pulse duration is determined and limited by the rather narrow optical bandwidth of the source as is well known for the mode-locking techniques of different laser materials.

Considering these limitations, instead of the use of these materials we suggested previously [1] another tentative approach which may exploit the fact that very wide optical spectra occur in the cases of multiple harmonic generation of atoms [2,3] and which may therefore result in the occurrence of a near attosecond light pulse sequence.

Meanwhile ref. [4] suggested that oscillations of six equidistant frequencies, which may be produced by sum- and frequency-mixing of light of two separate continuous lasers, may furnish phase controlled Fourier synthesis (with bandwidth $\hbar\Delta\omega \sim 2$ eV), i.e., short pulse durations of < 1 fs. The realization of this principle requires a complex system, containing two simultaneously working lasers, computer controlled electronics for ensuring the phase stabilizations, different nonlinear optical elements, special mechanical stability, etc.

In this Letter we describe and detail our idea [1] which may offer a far more simplified solution resulting in even shorter pulse durations. This principle is based on the multiple harmonic generation of atoms [2,3] induced by strong laser pulses. After the first experimental results [2] used in our calculation [1] further extended experimental investigations [5–7] and theoretical interpretations [8–11] have been performed for the process, the typical characteristics of which are the following (see also reviews [12,13]). When atomic beams of noble gases are illuminated with focussed Nd laser pulses of 30–40 ps duration above 10^{12} W/cm^2 intensities, odd harmonic photons ($n\hbar\omega$, $n=3, 5, 7, \ldots$) of the driving laser ($\hbar\omega = 1.17$ eV) are generated. According to the theory [8–11] at relatively high ($> 10^{13}$ W/cm^2) intensities the interaction is no more perturbative, therefore the spectrum has a special form in which after a rapid decrease at the first few harmonics there is a long plateau, which may extend up to high n values (e.g., $n=53$ for Ne atoms [14]) with a sharp cutoff, as has been verified experimentally [2,5–7,13,14]. That is the plateau which serves for the base of our idea.

According to the theory and observations in this non-perturbative region [13]:

(1) *All* Fourier components are emitted from the same (focal) region and propagate collinearly in the same single beam;

(2) The theories predict that the phases of the strictly equidistant stimulated harmonics are inherently fixed in time, which fact automatically ensures the fulfillment of the correct Fourier synthesis;

(3) The directional matching of the harmonic components along the plateau is surprisingly good in this strong field (non-perturbative) region.

Due to the experimentally found approximatively identical ($n=5$) laser intensity dependence order of each component of the plateau, the shortened pulse durations of the components are also roughly identical and equal to half the laser pulse. More or less the same is true for the far field spatial profiles of the harmonics: their preponderant central parts coincide well and have the same spot diameter and divergency values (much less than that of the laser).

Now, the overall effective spectral bandwidth is $\Delta\omega \sim 2N\omega$, where N is the number of odd harmonics of the plateau and $N=n_p-n_c$, where n_p and n_c are the first and last component of the plateau, respectively. Therefore the expected pulse duration after the Fourier synthesis is $\tau \sim 1/\Delta\omega \sim 1/2N\omega$. Using the experimental values [2] ($\omega \sim 1.8\times10^{15}\ \mathrm{s}^{-1}$ for a Nd laser, $N=9$ for xenon) a rough estimation immediately gives $\tau \sim 30\times10^{-18}\ \mathrm{s}=30$ as. The period of the inducing Nd laser light being $T=2\pi/\omega$, furthermore, the equidistant spacing between the harmonics being 2ω, the resulting beam will have a sequence of light peaks following each other by a period $\frac{1}{2}T$. The more precise characteristics as pulse shapes, durations, etc., are computed below. Due to the interference of the N roughly equal electric field amplitudes E_n, an N^2 times intensity increase is expected with respect to the intensity ($\sim E_n^2$) of an individual harmonic component E_n.

First we calculate the resulting electric fields values

$$E(t)=\sum_{n=n_p}^{n_c} E_n \cos(n\omega_0 t)\ ,$$

where $n_p\omega_0$ is the first and $n_c\omega_0$ is the last (cutoff) spectral component of the plateau. E_n is the electric field of the odd nth component ($n=1, 3, 5, \dots$). The E_n values may be determined from the experimental data [2,14], these E_n values are not rigorously equal, but their values are very close.

For the estimation of the intensity $I(t)$ of such pulses we cannot use routinely the usual intensity

$$I(t)=\frac{1}{T}\int E^2(t)\,\mathrm{d}t\ ,$$

the τ pulse duration being shorter than the laser oscillation time T. Therefore we simply calculate the resulting squared field strength $E^2(t)$ which characterizes well the instantaneous intensity (to which square detecting processes are sensitive) at each moment in time:

$$E^2(t)=\left(\sum_{n=n_p}^{n_c} E_n \cos(n\omega_0 t)\right)^2 .$$

To demonstrate the above mentioned principle, we calculated numerically the coherent (Fourier) sum of odd harmonics to determine both $E(t)$ and $E^2(t)$ for various gases.

From the figures of refs. [2] and [14] we may determine the relative electric field amplitudes E_n of the plateau regions. Identical time and spatial shapes for the harmonic beams are assumed [13] and the n-times higher photon energy of the nth harmonics is taken into account: $E_n \propto \sqrt{I_n} \propto \sqrt{nK_n}$, where I_n is the intensity, and K_n is the observed number of photons of the nth harmonics, respectively. On the basis of this consideration we may compute the functions $E(t)$ and $E^2(t)$. For illustrative purposes xenon was chosen (with the measured plateau between $n_p=5$ and $n_c=21$) as a typical example, the characteristic results of which are shown in fig. 1.

We can see that the resulting light consists of peaks following each other with period $\frac{1}{2}T$. The resulting pulse duration τ for E^2 is about 70 as. Each peak is accompanied by similar short-duration wings of 15% height relative to the main peaks. Similar results were obtained for all noble gases from the experiments which are summarized in table 1. We summarize in table 1 the half widths (FWHM) of the resulting light peaks in attoseconds both for the simple case $E(t)$ and for the case $E^2(t)$. The right column shows the increase of the maximum values of E^2 with respect to those relating to the individual multiharmonic components (E_n^2).

The estimated absolute value of the power and light intensity in each of these short light pulses is ~ 10 W and $10^4\ \mathrm{W/cm^2}$, respectively, using the experimental data of refs. [2,14].

As for the experimental realization, we have to keep

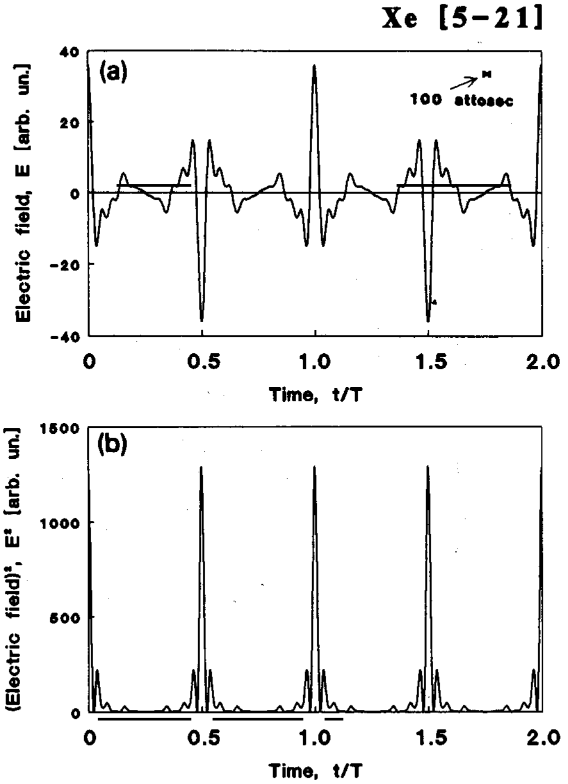

Fig. 1. The electric field strength $E(t)$ (a) and the square of the electric field strength $E^2(t)$ (b) of the light beam, in which high harmonic modes are coherently summarized from the 5th up to the 21st harmonic component. The results are plotted for two full oscillation periods $(2T)$ of the basic harmonic component ω_0 of the Nd laser beam $(T = 3.53$ fs).

Table 1
Resulting pulse durations and pulse height increase ratios after the Fourier synthesis of high harmonic components of the plateau region for various noble gases. The first (n_p) and the last (n_c) component of the plateau region are given

Gas	n_p	n_c	τ from $E(t)$ (as)	τ from $E^2(t)$ (as)	Increase ratio $(\Sigma E_n^2)/E_n^2$
xenon	5	21	97	72	81
krypton	5	25	76	56	169
argon	7	29	74	52	180
neon	13	53	38	28	441

in mind two important facts. First, the pulsed high order harmonics have very short wavelengths, therefore optical elements and manipulations as used in ref. [4] cannot be applied *at all*. On the other hand, the first few relatively strong harmonics (including that of the laser) before the plateau region might completely mask the observation of the short pulses in question produced by the Fourier synthesis of only the plateau components. Consequently, the suppression or reduction of these first few components is necessary. This reduction may be realized due to the fortunate good phase matching found experimentally [13,15] and treated widely in theoretical papers [12,13], according to which the divergencies and the spot sizes of the plateau harmonics are identical and much lower than those of the harmonics before the plateau. Therefore by inserting an appropriate diaphragm combination into the beam, the plateau components propagate unchanged, while the others having large spot sizes will be reduced in intensity by a factor 10–100, equalizing at the same time the values of all components for the Fourier synthesis.

It is clear that after presenting the computed characteristic of our suggested principle, in a real experiment all parameters and conditions (choice of gas, focussing control, space and time profiles, etc.) have to be optimized. Slight modifications of the presented idealized envelope $E^2(t)$ and τ values are expected from our more detailed preliminary computations to find these optimum values using more recent theoretical and experimental results.

The detection of this type of light containing a sequence of very sharp peaks with the repetition frequency of visible light necessitates a completely new method for observation. Due to the extremely short durations presented in table 1, this may possibly be realized only by atomic autocorrelation type methods.

In conclusion, we have described a proposal for the production of near-attosecond duration light pulses based on the multiple harmonic generation in gases. From recently published theoretical and experimental data our computations show that from this interaction the emission of a sequence of 30 as duration, $\sim 10^4$ W/cm^2 intensity pulses can be expected and realized in a compact simple experimental setup using single Nd laser pulses.

Finally we remark that the described idea and technique seems to be realizable in all nonlinear processes, where high harmonic generation takes place, e.g., in the case of the collinear harmonic generation on metal surfaces – in which case both odd and even harmonics appear – observed by us recently [16,17], in the case of multiphoton light scattering by free electrons, etc.

This work has been supported by the National Scientific Research Foundation (OTKA) of the Hungarian Academy of Sciences, Grant No. 2936.

References

[1] Gy. Farkas and Cs. Tóth, Natl. Sci. Res. Found. (OTKA) Report of Hung. Acad. Sci. 1783/89 (Budapest, 1989).
[2] M. Ferray, A. L'Huillier, X.F. Li, L.-A. Lompré, G. Mainfray and C. Manus, J. Phys. B 21 (1988) L31.
[3] A. McPherson, G. Gibson, H. Jara, U. Johann, T.S. Luk, I. McIntyre, K. Boyer and C.K. Rhodes, J. Opt. Soc. Am. B 4 (1987) 595.
[4] T.W. Hänsch, Opt. Commun. 80 (1990) 71.
[5] X.F. Li, A. L'Huillier, M. Ferray, L.A. Lompré, G. Mainfray and C. Manus, Phys. Rev. A 39 (1989) 5751.
[6] L.A. Lompré, A. L'Huillier, M. Ferray, P. Monot, G. Mainfray and C. Manus, J. Opt. Soc. Am. B 7 (1990) 754.
[7] N. Sarakura, K. Hata, T. Adachi, R. Nodomi, M. Watanabe and S. Watanabe, Phys. Rev. A 43 (1991) 1669.
[8] K.C. Kulander and B.W. Shore, Phys. Rev. Lett. 62 (1989) 524.
[9] J.H. Eberly, Q. Su and J. Javanainen, Phys. Rev. Lett. 62 (1989) 881.
[10] R.M. Potvliege and R. Shakeshaft, Phys. Rev. A 40 (1989) 3061.
[11] G. Bandarage, A. Maquet and J. Cooper, Phys. Rev. A 41 (1990) 1744.
[12] A. L'Huillier, K.J. Schafer and K.C. Kulander, Phys. Rev. Lett. 66 (1991) 2200.
[13] A. L'Huillier, K.J. Schafer and K.C. Kulander, J. Phys. 24 (1991) 3315.
[14] A. L'Huillier, L.A. Lompré, G. Mainfray and C. Manus, in: Multiphoton processes, eds. G. Mainfray and P. Agostini (CEA-Saclay, Gif sur Yvette, 1990) p. 45.
[15] A. L'Huillier, P. Balcou and L.A. Lompré, Phys. Rev. Lett. 68 (1992) 166.
[16] Gy. Farkas, Cs. Tóth, S.D. Moustaizis, N.A. Papadogiannis and C. Fotakis, submitted for publication; PWe016, at XVIII Int. Quantum Electronic Conf., IQEC'92, 14–19 June 1992.