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High-order harmonic generation in a narrow spectral range

V D Taranukhin

***Abstract*. It is proposed to use the two-component pumping of atoms by a strong low-frequency field and a high-frequency ultrashort pulse for the generation of high-order harmonics. For a fixed relative phase of the pumping fields, high-order harmonics are generated in a rather narrow spectral range. The interaction of atoms with the two-component field also determines the limiting efficiency of high-order harmonic generation. The effect can be reversed for the enhancement of high-order har-monics by using additional high-frequency pump radiation.**

Keywords: *ionisation of atoms, high-order harmonics, two-component pumping*.

The above-threshold tunnel ionisation of atoms enables one to control the ionisation moment (within the limits of the optical cycle) by the magnitude or direction of the drift velocity of a photoelectron incident on a photodetector [1]. In Ref. [2], it was proposed to use this effect for measuring the duration of femtosecond and attosecond pulses. In this scheme, the ionisation phase is specified by a high-frequency (HF) radiation pulse being measured, while the above-threshold gain of photoelectron energies is determined by the strong low-frequency (LF) field. In this paper, it is proposed to use the phase control of ionisation in the two-component field for high-order harmonic generation in a narrow spectral region [3].

Commonly, the spectrum of high-order harmonics represents a wide plateau, which spreads from the pump frequency to the frequency $\Omega \approx 3.17U_p + U_i$ [1], where U_p is the ponderomotive potential of pump radiation and U_i is the ionisation potential of an atom. However, only a certain part of this spectrum is needed for applications. Let us show that the two-component pumping enables one to substantially narrow the spectrum of high-order harmonics, thereby increasing the efficiency of the use of this spectrum.

The high-order harmonic generation includes three stages: (1) proper atomic ionisation (the transfer of a photoelectron to the continuum); (2) the gain of energy $\sim U_p$ by an electron (the above-threshold ionisation stage); and (3) electron recombination with a parent ion, which is accompanied by high-order harmonic generation of a photon with the frequency $\omega_n = kU_p + U_i$. The factor k varies from zero to 3.17 depending on the ionisation phase, which leads to the generation of a wide spectrum of high-order harmonics. The maximum lasing frequency Ω corresponds to the phase $\varphi \approx \pi/10$, which is calculated from the maximum of the pump field (see Fig. 1 below). Because of the strong nonlinearity of high-order harmonic generation, the 'fine structure' of this spectrum has a complex form. In the case of short pump pulses, the spectrum is continuous.

Consider the pumping by the two-component field, which is formed by a strong LF field (this field alone is unable to ionise an atom) and the HF field of ultrashort pulses with $\omega_H > U_i$, which is able to produce, for example, one-photon atomic ionisation and whose duration is considerably shorter than the optical cycle T of the LF field. We have

$$\boldsymbol{E} = \boldsymbol{E}_L \cos\omega_L t + \boldsymbol{E}_H \cos(\omega_H t + \varphi), \quad (1)$$

where t is the time; $\boldsymbol{E}_L \gg \boldsymbol{E}_H$ are the amplitudes; $\omega_L \ll \omega_H$ are the frequencies of LF and HF fields, respectively. Both fields are assumed to be linearly polarised along the x axis and synchronised in time, so that the HF pulse corresponds to a certain phase φ of the optical cycle of the LF field. If the amplitude E_L is insufficiently high for electron tunnelling from an atom, the atomic ionisation is governed by a short HF radiation pulse. As for the above-threshold stage, it is determined by the LF field.

Further calculations and estimates will be made for the following experimental conditions. The LF field is produced by a CO_2 laser with the radiation intensity $I_L = 6 \times 10^{13}$ W cm^{-2} (tunnel ionisation, for example, of He atoms by this radiation can be neglected, but the ponderomotive energy of this radiation is considerable: $U_p \approx 500$ eV), and the HF component represents radiation at the frequency $\omega_H \sim 25$ eV with duration $\tau \sim 1 - 3$ fs and intensity $I_H \sim 10^{10}$ W cm^{-2} (such radiation is produced in modern experiments on high-order harmonic generation [4]).

If $\omega_H \gtrsim U_i$, the photoelectron velocity at the moment it enters the continuum is $v_0 \approx 0$, more precisely, $0 < v_0 < v_{0m}$. The maximum initial photoelectron velocity in the absence of phase modulation of the HF pulse is $v_{0m} = (2\hbar/m\tau)^{1/2}$, and it is determined by the pulse duration τ (m is the electron mass). In this case, the ionisation probability is proportional to the intensity I_H and the threshold cross section for one-photon ionisation. The further evolution of the photoelectron wave packet proceeds under the action of the LF field and can be described by the classical trajectory $x(t)$ of the packet centre.

Neglecting the Coulomb attraction to the parent ion (which is valid for $U_p \gg U_i$), one can write the equation of this trajectory in the simple form

$$\frac{d^2x}{dt^2} = \frac{e}{m} E_L \cos(\omega_L t + \varphi), \quad (2)$$

V D Taranukhin International Teaching and Research Laser Centre, M V Lomonosov Moscow State University, Vorob'evy gory, 119899 Moscow, Russia

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where e is the electron charge. The solution of this equation with the initial conditions $x(t=0)=0, v(t=0)=v_0$ allows one to calculate the kinetic energy ε_k of an electron at the moment of its return to the parent ion (i.e., to the point $x=0$, where recombination takes place). In this case, the packet spread, which is not described by Eqn (2), affects the electron recombination efficiency and has no effect on the electron kinetic energy ε_k .

The energy ε_k is determined by the initial electron velocity v_0 and the ionisation phase φ (Fig. 1). The latter is fixed by the HF field (the phases $\varphi_{1,2}$ in Fig. 1 correspond to the onset and the end of the HF pulse), which allows one to control the frequency of recombination emission, i.e., the spectrum of high-order harmonics. Note that the return of a photoelectron to the parent ion and its recombination occur only then when the velocity v_0 and the LF field have opposite directions. Because one-photon ionisation produces a symmetric two-lobe wave packet ($\pm v_0$), half the photoelectrons give no contribution to high-order harmonic generation.

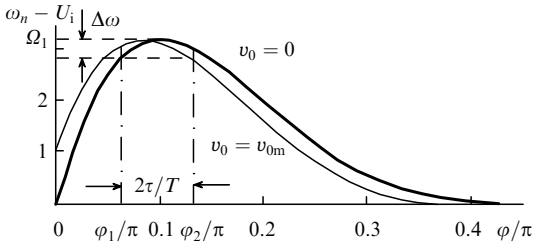

Figure 1. Dependence of the recombination emission frequency on the ionisation phase (calculated from the maximum of the LF field) for different initial photoelectron velocities. The gating of the spectrum of high-order harmonics by a HF radiation pulse with duration τ is shown.

The calculation of electron energy ε_k (actually, the spectrum of high-order harmonics $\omega_n = \varepsilon_k + U_i$) by Eqn (2) is illustrated in Fig. 1. One can see that the HF pulse strobes ionisation phases ($\varphi_1 < \varphi < \varphi_2$), resulting in the generation of high-order harmonics only in a narrow spectral range $\Delta\omega$. The contribution to $\Delta\omega$ is made only by those components that are found between the curves $\varepsilon_k(\varphi)$ for $v_0 = 0$ and $\varepsilon_k(\varphi)$ for $v_0 = v_{0m}$ and correspond to the ionisation phases $\varphi_1 < \varphi < \varphi_2$. Fig. 1 also illustrates the generation of high-order harmonics in the narrow frequency range $\Delta\omega \approx 0.12\Omega_1$ (where $\Omega_1 = \Omega - U_i$) by a HF pulse with duration $\tau = (\varphi_2 - \varphi_1)T/2\pi \approx 1.2$ fs and relative phase $\varphi \approx \pi/10$. One can tune the central frequency of this spectrum by varying the LF field amplitude: $\Omega_1 \sim I_L$. The velocity v_{0m} used in Fig. 1 corresponds to a HF pulse with a certain phase modulation. For the HF pulse with the same duration ($\tau = 1.2$ fs), but without phase modulation, the spectrum of high-order harmonics is narrower by approximately 30 %.

A decrease in the HF pulse duration results in an increase in the initial electron velocity v_{0m}, which (for sufficiently small τ) may cause broadening of the spectrum of high-order harmonics rather than its narrowing. Assuming that the dependence $\varepsilon_k(\varphi)$ near the top of the curve is parabolic and the HF pulse has no phase modulation, we find from Eqn (2)

$$\Delta\omega/\Omega_1 = \alpha(\tau + \beta/\tau^{1/2})^2. \quad (3)$$

By expressing T and τ in femtoseconds and Ω_1 in electronvolts, we obtain $\alpha \approx 55.8/T^2$, $\beta \approx 0.17T/\Omega_1^{1/2}$. It follows from (3) that the minimum width of the spectrum of high-order harmonics

$$\Delta\omega_{min} = 9\alpha\tau_0^2\Omega_1 \quad (4)$$

is realised for the HF pulse duration $\tau_0 = (\beta/2)^{2/3} \sim T^{2/3}\Omega_1^{-1/3}$. For $T \approx 35$ fs (LF radiation of a CO_2 laser), numerical estimates give $\tau_0 \approx 0.2$ fs and $\Delta\omega_{min}/\Omega_1 \approx 0.016$. The absolute width of the spectrum increases with increasing LF radiation intensity: $\Delta\omega_{min} \sim \Omega_1^{1/3}$, $\Omega_1 \sim I_L$. However, the relative width of the spectrum decreases with increasing intensity I_L ($\Delta\omega_{min}/\Omega_1 \sim \Omega_1^{-2/3}$) and may become noticeably smaller than 1 %.

Thus, to obtain an ultimately narrow spectrum of high-order harmonics, one should use HF pulses of optimum duration without phase modulation and synchronise them with the LF field near the optimum phase $\varphi \leqslant \pi/10$ (the higher is the maximum initial electron velocity v_{0m}, the closer should be the HF pulse to the maximum of the LF field).

In conclusion, note that the mechanism of atomic ionisation by the two-component field, which was analysed here, also explains the absorption of harmonics in a usual (one-component) scheme of high-order harmonic generation. In this case, harmonics may be involved in the tunnel ionisation of an atom by the LF pump field by changing the effective atomic ionisation potential: $U_{ief} = U_i - \omega_H$ [5]. Because of the exponential dependence of the probability of tunnel ionisation on the ionisation potential, this leads to a sharp (by several orders of magnitude) increase in the ionisation rate and efficient absorption of high-order harmonics. This effect restricts the limiting efficiency of high-order harmonic generation in the case when the absorption length of high-order harmonics is smaller than the length of an active medium or the coherence length [4].

However, this effect can be used for a substantial increase in the efficiency of high-order harmonic generation by using additional HF pumping. For instance, if a relatively weak radiation at the frequency of one of the harmonics is used in addition to the main pump radiation, the rate of tunnel ionisation of atoms considerably increases. This will lead, in turn, to an increase in the intensities of all components of the spectrum of high-order harmonics. There will be no real enhancement of radiation at the frequency of the harmonic that is used for additional pumping of atoms (because of a low efficiency of high-harmonic generation), but the remaining harmonics of the spectrum will be noticeably enhanced.

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