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Atomic photoeffect in the pulsed fields of a high-order harmonic and the fundamental wave

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***Abstract*. The atomic photoeffect is studied in the pulsed fields of two coherent waves: a high-order harmonic wave and the fundamental wave. Expressions are derived for the intensity of the main peak and satellites in the photoelectron energy spectrum for an arbitrary delay between the pulses. The role of inelastic Coulomb rescattering of photoelectrons from the residual ion in the fundamental wave field, in the production of satellites is analysed.**

Keywords: *photoeffect, Coulomb rescattering, high-order harmonics, pulsed fields.*

1. Introduction

High-order harmonic generation in high-power laser field – atom interactions has been adequately studied, both experimentally and theoretically. From the practical viewpoint, the generation of high-order harmonics of pulsed IR laser radiation makes it possible to produce high-intensity femtosecond pulses in the near- and far-UV ranges. Two problems are currently of interest: the measurement of harmonic formation duration and the measurement of the relative phase of the fields of two neighbouring harmonics, which is critical for efficient generation. The former problem is of general physical significance and the latter is of practical significance, because its solution is related to the feasibility of investigation of attosecond high-order harmonic pulse generation [1].

Because of the extremely short duration of harmonic generation, the only technique that enables its experimental evaluation is a cross-correlation experiment [2 – 4]. The laser fundamental wave is split into two beams. One of them (the higher-power beam) is employed as the pump wave to generate high-order harmonics, which are then directed to the gas medium. The second, relatively weak, pulse of the fundamental wave is also directed to the same medium. Therefore, the atomic photoeffect in the gas medium takes place in the presence of two pulses: the high-order harmonic wave and the fundamental wave. Apart from the main photoionisation peak, the photoelectron spectrum emerging in this case also contains satellites, which are separated from the main peak by a distance equal to the fundamental-wave photon energy.

The amplitude of the satellites depends, in particular, on the pulse durations and the time delay between them. This dependence is exponential, the exponent being determined by the ratio of pulse durations. The investigation of this dependence under the conditions when the pulse duration of the fundamental wave and the time delay are controllable underlies the cross-correlation method for determining the high-order harmonic pulse duration.

The dependence of satellite amplitude on the time delay is commonly described using a phenomenological approach [2 – 4]. However, this approach does not permit, in particular, determining the angular photoelectron distribution in the satellites. Nor is it possible to determine the pre-exponential factor as a function of the amplitude and time delay. Note also that the results of the above-mentioned papers, wherein pump waves of different intensity were employed for harmonic generation, contradict each other to a great extent.

In this paper we develop a consistent quantum-mechanical theory of the atomic photoeffect in the fields of two coherent pulsed waves – the high-order harmonic and fundamental waves – with an arbitrary time delay between them.

2. Formulation of the problem. Basic equations

The cross-correlation method for determining the duration of a high-order harmonic pulse relies on the above-threshold ionisation of atoms in the fields of two coherent pulsed waves: the high-order harmonic wave with a frequency Ω_s and the fundamental wave with a frequency ω ($\Omega_s = s\omega$, where s is the harmonic number). The medium in cross-correlation experiments consists of rare-gas atoms (He, Ar) with a high ionisation potential.

The fundamental wave intensity is assumed to be moderate ($I_{10} \sim 10^{11} - 10^{12}$ W cm^{-2}), when the Keldysh adiabaticity parameter [5] $\gamma \approx 10$ and the multiphoton ionisation of the atoms by the field of this wave can be neglected for a short pulse duration ($\tau_1 \sim 100$ fs – 1 ps). On the other hand, the high-order harmonic photon energy should be high enough ($\Omega_s \approx 30 - 50$ eV) for the single-photon ionisation of the atoms to occur with an appreciable probability under harmonic field irradiation. The simultaneous presence of the two waves in the interaction volume

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results in the production of the main peak with an energy $\varepsilon_{0s} = -(|E_0| + U_p) + \Omega_s$ and the satellites with energies $\varepsilon_{pn} = \varepsilon_{0s} \pm n\omega$, in the photoelectron spectrum, where $|E_0| + U_p$ is the electron binding energy in the atomic ground state with the inclusion of the ponderomotive potential U_p in the fundamental wave field, and n is the satellite order (below, we consider first-order satellites, when $n = 1$)*.

The atom–wave fields interaction is described using a ***pA*** gauge, in which the operators

$$\hat{V}_1 = \mathrm{i}\frac{\mathrm{e}A_{10}(\boldsymbol{e}_1\nabla)}{2m_\mathrm{e}}\{\exp[\mathrm{i}(\omega t - \boldsymbol{kr})] + \mathrm{c.\,c.}\}f_1(t), \tag{1}$$

$$\hat{V}_2 = \mathrm{i}\frac{\mathrm{e}A_{20}(\boldsymbol{e}_2\nabla)}{2m_\mathrm{e}}\exp[\mathrm{i}(\Omega_s t - \boldsymbol{Kr})]f_2(t) \tag{2}$$

are considered, where $\boldsymbol{k}$ and $\boldsymbol{K}$ are the wave vectors of the fundamental wave and the high-order harmonic wave; $\boldsymbol{A}_{10}$ and $\boldsymbol{A}_{20}$ are the amplitudes of the vector potential of the corresponding waves with unit polarisation vectors $\boldsymbol{e}_1$ and $\boldsymbol{e}_2$ (we assume below that $\boldsymbol{e}_1 = \boldsymbol{e}_2 \equiv \boldsymbol{e}$); $f_1(t) = \exp[-(t-\Delta\tau)^2/\tau_1^2]$ and $f_2(t) = \exp(-t^2/\tau_2^2)$ are the time envelopes of the wave amplitudes, whose peaks are shifted by the time delay $\Delta\tau$.

Note that there exists another interaction gauge, which is called the ***dE*** approximation. However, in Ref. [6] these two gauges were shown to be equivalent in the dipole approximation used in our work. The adiabatic approximation during the onset and termination of the fundamental wave ($\omega\tau_1 \gg 1$) is assumed to be fulfilled. It permits separating the rapidly oscillating factors in the electron wave functions of the continuous spectrum.

The operator $\hat{V}_{12} \sim \boldsymbol{A}_1\boldsymbol{A}_2$ plays an insignificant part in satellite production and is therefore neglected in our calculations (see the estimates below). The operator $\hat{V}_2$ is used in the first order of the perturbation theory when calculating the probability amplitude of single-photon atom ionisation by the high-order harmonic field, and the operator $\hat{V}_1$ is used when constructing the basis for electron wave functions of the continuum in the field of a strong wave (the Keldysh method [5]).The inclusion of Coulomb interaction between the photoelectron and the residual ion calls for a separate consideration and is discussed in greater detail below in Section 4.

Note that there exist two satellite production mechanisms. The satellites can appear both due to direct atom photoionisation by the high-order harmonic field when the effect of the strong fundamental wave is included, and due to a stepwise transition with the initial harmonic-induced atom ionisation followed by the inelastic Coulomb photoelectron rescattering from the residual ion with a capture (emission) of the fundamental-wave photon. In principle, more complex versions of stepwise electron transitions to the final satellite states are also possible through Coulomb rescattering. However, these transitions arise when higher orders of the perturbation theory are included. When the fundamental-wave intensity is not too high, the contribution of the above processes to the probability amplitude of satellite production can be neglected.

The probability amplitude of direct atom photoionisation in the presence of both waves is given by the expression

$$A_{\boldsymbol{p}}(\infty) = -\mathrm{i}\int_{-\infty}^{\infty}\mathrm{d}t\langle\Psi_{\boldsymbol{p}}^*|\hat{V}_2|\Psi_0\rangle, \tag{3}$$

where Ψ_0 is the wave function of the atomic ground state (the 1s-state function of a hydrogen-like ion is taken in specific calculations of the matrix elements);

$$\Psi_{\boldsymbol{p}}(\boldsymbol{r}, t) = \exp[-\mathrm{i}(\varepsilon_p t - \boldsymbol{pr})]\exp(\mathrm{i}y\sin\varphi) \tag{4}$$

is the electron wave function of the continuum in the fundamental wave field (the Volkov wave); $y = e(\boldsymbol{A}_1\boldsymbol{p})/(m_\mathrm{e}\omega) \equiv y_0 f_1(t)$; $y_0 = e(\boldsymbol{A}_{10}\boldsymbol{p})/(m_\mathrm{e}\omega)$; $\varphi = \omega t - \boldsymbol{kr}$; and ε_p is the energy of a photoelectron with the momentum $\boldsymbol{p}$. The function $\Psi_{\boldsymbol{p}}$ is then expanded into a series in terms of the eigen-energy states (the well-known expansion in Bessel functions [7]). The argument of the Bessel functions is small for typical parameter values in cross-correlation experiments: $(y_0/2) < 1$, and the results of our work were obtained in lowest-order approximations in this parameter.

The expression for the probability amplitude of direct atom photoionisation (3) was derived by neglecting the interaction of an electron in the continuum with the Coulomb ion potential. However, it follows from cross-correlation experimental data that the photoelectron energies are, as a rule, moderate: $\varepsilon_p \leqslant 10$ eV. That is why the inelastic Coulomb rescattering of the photoelectrons by ions can make a significant contribution to the satellite intensities.

In the first order in the Coulombian interaction, the probability amplitude of satellite formation is given by the expression

$$\tilde{A}_{\boldsymbol{p}}(\infty) = (-\mathrm{i})^2\int\frac{\mathrm{d}\boldsymbol{p}'}{(2\pi)^3}\int_{-\infty}^{\infty}\mathrm{d}t\langle\Psi_{\boldsymbol{p}}^*|\hat{V}_\mathrm{c}|\Psi_{\boldsymbol{p}'}\rangle \times\int_{-\infty}^{t}\mathrm{d}t_1\langle\Psi_{\boldsymbol{p}'}^*|\hat{V}_2|\Psi_0\rangle, \tag{5}$$

where $\Psi_{\boldsymbol{p}'}$ is the wave function of an electron of the continuum in the intermediate state with $\varepsilon_{\boldsymbol{p}'}, \boldsymbol{p}'$; $\langle\Psi_{\boldsymbol{p}}^*|\hat{V}_\mathrm{c}|\Psi_{\boldsymbol{p}'}\rangle$ is the Coulomb interaction matrix element, which depends on the transferred momentum $\Delta\boldsymbol{p} = \boldsymbol{p} - \boldsymbol{p}'$.

3. Probability amplitude of direct atom photoionisation

The calculation of the matrix element of the operator $\hat{V}_2$ in the dipole approximation ($p \gg sk$) and subsequent integration in expression (3) lead to the expression for the probability amplitude of direct atom photoionisation with a photoelectron in the main peak ($\varepsilon_p = \varepsilon_{0s}$, where $\varepsilon_p = \varepsilon_{pn}$ for $n = 0$):

$$A^{(\varepsilon_p=\varepsilon_{0s})} = 4\sqrt{\pi}\alpha\tau_2 eA_{20}(\pi a_0^3)^{1/2}\left(\frac{\mathrm{Ry}}{I_0}\right)^{3/4}\frac{(\varepsilon_p/I_0)^{1/2}}{(1+\varepsilon_p/I_0)^2} \times\exp[-(\varepsilon_p - \varepsilon_{0s})^2\tau_2^2/4]\cos\theta_0, \tag{6}$$

where α is the fine structure constant; a_0 is the first Bohr radius of a hydrogen atom; Ry is the Rydberg constant; $I_0 = |E_0|$ is the binding energy of the atomic ground state;

*In our paper, the system of units is used in which $\hbar = c = 1$.

and θ_0 is the angle between the directions of photoelectron momentum $\boldsymbol{p}$ and wave polarisation.

The exponential in expression (6) describes the photoelectron energy spectrum in the neighbourhood of the main peak, which arises when the atom absorbs a photon with a frequency Ω_s. We draw attention to the fact that expression (6) was obtained in the zero approximation for the electron–fundamental wave interaction [a plane wave approximation in expression (4)]. It is evident that this approximation is insufficient to find, with the aid of operator $\hat{V}_2$ from formula (3), the expression describing the electron transition to the satellites. However, even in the first approximation in the parameter y_0 of the expansion of expression (4), it is possible to obtain from formula (3) the expression for the probability amplitude of direct atom ionisation with the formation of satellites (the photoelectron energy is $\varepsilon_p = \varepsilon_{0s} \pm \omega$, where $\varepsilon_p = \varepsilon_{pn}$ for $n = 1$):

$$A^{(\varepsilon_p=\varepsilon_{0s}\pm\omega)} = \sqrt{2\pi}\alpha\tau_1\left(\frac{x}{1+x}\right)^{1/2} eA_{20}\frac{eA_{10}}{\omega}(\pi a_0^3)^{1/2}$$

$$\times\left(\frac{I_0}{m_e}\right)^{1/2}\left(\frac{\mathrm{Ry}}{I_0}\right)^{3/4}\frac{(\varepsilon_p/I_0)^{1/2}}{(1+\varepsilon_p/I_0)^2}\exp\left[-\frac{(\Delta\tau/\tau_1)^2}{1+x}\right]$$

$$\times\exp\{-[\varepsilon_p-(\varepsilon_{0s}\pm\omega)]^2\tau_1^2 x/[4(1+x)]\}\cos^2\theta_0, \quad (7)$$

where $x \equiv \tau_2^2/\tau_1^2$ is a dimensionless parameter, which should be derived from experimental data for the determination of the duration of high-order harmonic generation.

Note that the parameter x appears both in the exponent, which describes the decay of the amplitude (7) with time delay $\Delta\tau$, and in the pre-exponential factor. The exponential itself arises in the natural way in the calculation of the Fourier transform of the overlap function $f_1(t)f_2(t)$ of the wave envelopes and coincides with the phenomenological result of Refs [2–4].

The expression for direct atom photoionisation with the formation of satellites in the final state can also be obtained [in the plane wave approximation in expression (4)] using the operator $\hat{V}_{12} = e^2(\boldsymbol{A}_1\boldsymbol{A}_2)$ from a formula similar to formula (3), by replacing $\hat{V}_2$ by $\hat{V}_{12}$. Indeed, the time dependence $\hat{V}_{12}(t)$ contains the combination of wave frequencies required for atom photoionisation to the satellite states. However, the ratio between the amplitude obtained employing the operator $\hat{V}_{12}$ and the amplitude (7) is determined by the quantity $\sim\omega/\varepsilon_p < 1$ and suggests that that the contribution of $\hat{V}_{12}$ to the satellite formation probability amplitude can be neglected in this case.

4. Probability amplitude of atomic photoeffect with the inclusion of Coulomb photoelectron rescattering from the residual ion

If we restrict ourselves to the process in which a photoelectron captures one photon of the fundamental wave, then by expanding function (4) in parameter y_0 in the lowest order, we obtain from (5)

$$\tilde{A}^{(\varepsilon_p=\varepsilon_{0s}\pm\omega)} = -\int\frac{\mathrm{d}\boldsymbol{p}'}{(2\pi)^3}\int_{-\infty}^{\infty}\mathrm{d}t\frac{\Delta y_0}{2}V_c(\Delta\boldsymbol{p})V_2(\boldsymbol{p}')$$

$$\times\exp\{\mathrm{i}[\varepsilon_p-(\varepsilon_{p'}\pm\omega)]t\}f_1(t)F(t), \quad (8)$$

where $V_c(\Delta\boldsymbol{p})$ and $V_2(\boldsymbol{p}')$ are the matrix elements of the corresponding operators;

$$F(t) = \int_{-\infty}^{t}\mathrm{d}t_1\exp[\mathrm{i}(\varepsilon_{p'}-\varepsilon_{0s})t_1]f_2(t_1). \quad (9)$$

The stepwise process responsible for the amplitude (8) is shown in Fig. 1. The integrand (8) containes the product $f_1(t)F(t)$, and therefore the main contribution to the amplitude $\tilde{A}^{(\varepsilon_p=\varepsilon_{0s}\pm\omega)}$ is made by the function $F(t)$ in the domain $|t| \leqslant \tau_1$ in the neighbourhood of $t \approx \Delta\tau$. This circumstance permits reducing the integral (9) to a tabular one [7] by the change of variable $t_1 = t + t'$. We perform calculations to obtain for $F(t)$ the expression

$$F(t) = \frac{\sqrt{\pi}}{2}\tau_1\sqrt{x}\exp\left[-\frac{(\varepsilon_{p'}-\varepsilon_{0s})^2\tau_1 x}{4}\right]$$

$$\times\left\{1+\Phi\left[\frac{t}{\tau_1\sqrt{x}}-\mathrm{i}\frac{(\varepsilon_{p'}-\varepsilon_{0s})\tau_1\sqrt{x}}{2}\right]\right\}, \quad (10)$$

where $\Phi(u)$ is the probability integral [7]. The time dependence of the function $F(t)$ is contained in the real part of the argument u of the probability integral; the imaginary part of the argument coincides with the exponent, which describes the energy spectrum $\varepsilon_{p'}$ of the intermediate electron state.

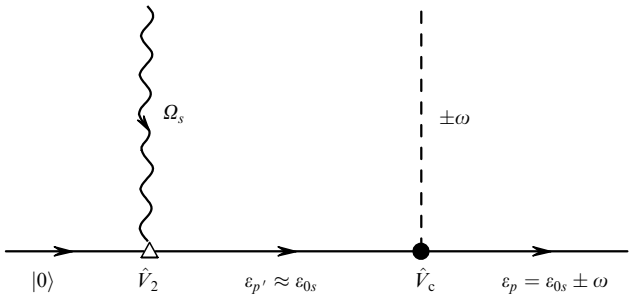

Figure 1. Feynman diagram describing the atomic photoeffect. The summit, which is represented as a triangle with a wavy line, corresponds to the interaction of an atom in the ground state $|0\rangle$ with the high-order harmonic field (the operator V_2) in the presence of the fundamental wave, resulting in the electron transition to the intermediate state with an energy $\varepsilon_{p'}$ in the continuum; the summit depicted with a dashed line with a circle (the operator V_c) describes the inelastic photoelectron rescattering by the Coulomb potential of the residual ion with the capture (emission) of one photon of the fundamental wave, which transfers the electron to the final state (the satellites with energies $\varepsilon_p = \varepsilon_{0s} \pm \omega$).

The subsequent calculations of the amplitude (8) are reduced to the substitution of (10) into (8) with the subsequent determination of the time interval by the method of steepest descent [8]. The calculation is described in detail in Appendix 1, and the result obtained is

$$\tilde{A}^{(\varepsilon_p=\varepsilon_{0s}\pm\omega)} = \frac{1}{\pi}(\mathrm{Ry}\,\tau_1)^2\sqrt{x}\,\frac{eA_{10}}{\omega}\frac{eA_{20}}{m_e}(\pi a_0^3)^{1/2}\times$$

$$\times\left(\frac{\mathrm{Ry}}{I_0}\right)^{3/4}\int_0^\infty \mathrm{d}z\,\frac{z}{(1+\mathrm{Ry}\,z/I_0)^2}\left[1+\Phi\left(\frac{t_\mathrm{c}-t_0}{\tau_1\sqrt{x}}\right)\right]$$

$$\times\exp[-(z-z_0)^2\mathrm{Ry}^2\tau_1^2x/4]\exp\{\mathrm{i}[\bar{z}_0-(z\pm q)]\mathrm{Ry}\,\Delta t\}$$

$$\times\exp\{-[\bar{z}_0-(z\pm q)]^2\mathrm{Ry}^2\tau_1^2/4\}J_{\theta\varphi} \qquad (11)$$

(for the notation, see Appendix 1). After cumbersome although easy calculations of tabulated integrals in expression (A1.5), we obtain

$$J_{\theta\varphi}=-\frac{\pi z^{1/2}}{4(\bar{z}_0z)^{3/2}}[F_0(z)+F_1(z)\cos^2\theta_0], \qquad (12)$$

where

$$F_0(z)=2(\bar{z}_0z)^{1/2}(\bar{z}_0+z)-(\bar{z}_0-z)^2\ln|v|;$$

$$v=(\sqrt{\bar{z}_0}+\sqrt{z})/(\sqrt{\bar{z}_0}-\sqrt{z});$$

$$F_1(z)=2[2(\bar{z}_0z)^{1/2}(\bar{z}_0-3z)-(\bar{z}_0^2+2\bar{z}_0z-3z^2)\ln|v|]. \qquad (13)$$

Substituting (12) into (11) leads to the following expression for the amplitude of the process under consideration:

$$\tilde{A}^{(\varepsilon_p=\varepsilon_{0s}\pm\omega)}=\frac{(\mathrm{Ry}\,\tau_1)^2}{4}\sqrt{x}\,\frac{eA_{10}}{\omega}\frac{eA_{20}}{m_\mathrm{e}}(\pi a_0^3)^{1/2}\left(\frac{\mathrm{Ry}}{I_0}\right)^{3/4}$$

$$\times\int_0^\infty \mathrm{d}z\,\frac{[F_0(z)+F_1(z)\cos^2\theta_0]}{(1+\mathrm{Ry}\,z/I_0)^2\bar{z}_0^{3/2}}\{1+\Phi[u(z)]\}$$

$$\times\exp\{\mathrm{i}[\bar{z}_0-(z\pm q)]\mathrm{Ry}\,\Delta t\}\exp\{-[\bar{z}_0-(z\pm q)]^2\mathrm{Ry}^2\tau_1^2/4\}$$

$$\times\exp[-(z-z_0)^2\mathrm{Ry}^2\tau_1^2x/4]. \qquad (14)$$

The integral in expression (14) is also calculated by the method of steepest descent, and the details of this calculation are given in Appendix 2. After the substitution of (A2.4) into expression (14), we find the final expression for the probability amplitude of satellite formation:

$$\tilde{A}^{(\varepsilon_p=\varepsilon_{0s}\pm\omega)}=\sqrt{\pi}\mathrm{Ry}\,\tau_1\left(\frac{x}{1+x}\right)^{1/2}\frac{eA_{10}}{\omega}\frac{eA_{20}}{m_\mathrm{e}}(\pi a_0^3)^{1/2}$$

$$\times\left(\frac{\mathrm{Ry}}{I_0}\right)^{3/4}\frac{F_0(\bar{z}_0)+F_1(\bar{z}_0)\cos^2\theta_0}{(1+\mathrm{Ry}\,z_0/I_0)^2\bar{z}_0^{3/2}}\exp\left\{-[\bar{z}_0-(z_0\pm q)]^2\right.$$

$$\left.\times\mathrm{Ry}^2\tau_1^2\frac{x}{4(1+x)}\right\}\exp\left[-\left(\frac{\Delta\tau}{\tau_1}\right)^2\frac{1}{1+x}\right]. \qquad (15)$$

Comparison of expressions (7) and (15) shows, in particular, that the corresponding amplitudes decay similarly with increasing time delay $\Delta\tau$, irrespective of the satellite formation mechanism.

5. Number of photoelectrons emitted by an atom per one pulse

We give formulas for the number of photoelectrons emitted by an atom per one pulse into the main peak ($\varepsilon_p=\varepsilon_{0s}$) and the satellites ($\varepsilon_p=\varepsilon_{0s}\pm\omega$). These formulas are obtained in a conventional way by integrating the squared modulus of the amplitudes (6), (7), and (15) taking into account the statistical weights of electrons in the final state. The final results are given in the ordinary system of units.

The expression for the number of photoelectrons emitted by the atom per pulse within a unit solid angle Ω_p into the main peak in the case of direct ionisation by the high-order harmonic field follows from expression (6) and has the form

$$\frac{\mathrm{d}N^{(\varepsilon_p=\varepsilon_{0s})}}{\mathrm{d}\Omega_p}=\left(\frac{2}{\pi}\right)^{1/2}\frac{\mathrm{Ry}\,\tau_1}{\hbar}\left(\frac{eA_{20}}{m_\mathrm{e}c^2}\right)^2\frac{m_\mathrm{e}c^2}{I_0}$$

$$\times\frac{(\varepsilon_{0s}/I_0)^{3/2}}{(1+\varepsilon_{0s}/I_0)^4}\cos^2\theta_0. \qquad (16)$$

The corresponding expression for the number of photoelectrons that make up satellites due to direct ionisation in the fields of two waves is derived from expression (7) and is

$$\frac{\mathrm{d}N^{(\varepsilon_p=\varepsilon_{0s}\pm\hbar\omega)}}{\mathrm{d}\Omega_p}=\left(\frac{2}{\pi}\right)^{1/2}\frac{\mathrm{Ry}\,\tau_1}{\hbar}\left(\frac{x}{1+x}\right)^{1/2}\left(\frac{eA_{20}}{m_\mathrm{e}c^2}\right)^2$$

$$\times\left(\frac{eA_{10}}{\hbar\omega}\right)^2\frac{(\varepsilon_{0s}/I_0)^{5/2}}{[1+\varepsilon_{0s}(1\pm\tilde{t})I_0]^4}(1\pm\tilde{t})^{5/2}$$

$$\times\exp\left[-\left(\frac{\Delta\tau}{\tau_1}\right)^2\frac{2}{1+x}\right]\cos^4\theta_0, \qquad (17)$$

where $\tilde{t}=\hbar\omega/\varepsilon_{0s}$ is a dimensionless parameter ($0\leqslant\tilde{t}\leqslant1$); the plus sign in expression (17) corresponds to the right satellite with an energy $\varepsilon_p=\varepsilon_{0s}+\hbar\omega=\varepsilon_{0s}(1+\tilde{t})$ and the minus sign to the left satellite with an energy $\varepsilon_p=\varepsilon_{0s}(1-\tilde{t})$.

The expression for the number of photoelectrons that form satellites and are emitted into a unit solid angle can be derived from expression (15) and is

$$\frac{\mathrm{d}\tilde{N}^{(\varepsilon_p=\varepsilon_{0s}\pm\hbar\omega)}}{\mathrm{d}\Omega_p}\propto\int_0^\infty\frac{[F_0(\bar{z}_0)+F_1(\bar{z}_0)\cos^2\theta_0]^2}{\bar{z}_0^{5/2}}$$

$$\times\exp\left\{-[\bar{z}_0-(z_0\pm q)]^2\mathrm{Ry}^2\tau_1^2\frac{x}{2(1+x)}\right\}\mathrm{d}\bar{z}_0. \qquad (18)$$

The result of integration depends on the location of the main photoelectron peak relative to the ionisation threshold. When the final photoelectron energy is high enough, when $\varepsilon_{0s}>\hbar\omega$ (the parameter $z_0\pm q>0$) and the saddle point of the phase of the exponent in expression (18) resides within the range of integration over the final photoelectron energy z_0, from expression (18) we obtain

$$\frac{\mathrm{d}\tilde{N}^{(\varepsilon_p=\varepsilon_{0s}\pm\hbar\omega)}}{\mathrm{d}\Omega_p}=\left(\frac{2}{\pi}\right)^{1/2}\frac{\mathrm{Ry}\,\tau_1}{\hbar}\left(\frac{x}{1+x}\right)^{1/2}\left(\frac{eA_{20}}{m_\mathrm{e}c^2}\right)^2\left(\frac{eA_{10}}{\hbar\omega}\right)^2$$

$$\times\frac{(\varepsilon_{0s}/I_0)^{3/2}}{(1+\varepsilon_{0s}/I_0)^4}\frac{[\Phi^{(\pm)}(\tilde{t})]^2}{(1\pm\tilde{t})^{5/2}}\exp\left[-\left(\frac{\Delta\tau}{\tau_1}\right)^2\frac{2}{1+x}\right], \qquad (19)$$

where

$$\Phi^{(\pm)}(\tilde{t})=F_0^{(\pm)}(\tilde{t})+F_1^{(\pm)}(\tilde{t})\cos^2\theta_0=(1\pm\tilde{t})^{1/2}(1\pm\tilde{t}/2)$$

$$-(\tilde{t}/2)^2\ln\left|\frac{(1\pm\tilde{t})^{1/2}+1}{(1\pm\tilde{t})^{1/2}-1}\right|-2\left[(1\pm\tilde{t})^{1/2}(1\mp\tilde{t}/2)\right.$$

$$\left.\pm\tilde{t}(1\pm\tilde{t}/4)\ln\left|\frac{(1\pm\tilde{t})^{1/2}+1}{(1\pm\tilde{t})^{1/2}-1}\right|\right]\cos^2\theta_0 \qquad (20)$$

and it is implied that $t < 1$.

We make a comment concerning expression (19). Strictly speaking, the formula for satellite intensities resulting from the integration with respect to $\bar{z}_0$, is expressed in terms of parabolic cylinder functions $D_\nu(w)$ [7]. However, for $\varepsilon_{0s} > \hbar\omega$ the argument w of these functions is large in the adiabatic approximation ($\omega\tau_1 \gg 1$), making it possible to resort to the well-known asymptotic representation for the functions D_ν [7]. This procedure is equivalent to employing in expression (18) the δ approximation for the exponent containing $\bar{z}_0$. The result of calculations is expression (19), which is universal in character and from which there follows a formula for satellite intensities in the case of high-lying photoelectron peaks for $\tilde{t} \approx 0$:

$$\frac{\mathrm{d}\tilde{N}^{(\varepsilon_p=\varepsilon_{0s}\pm\hbar\omega)}}{\mathrm{d}\Omega_{\boldsymbol{p}}} = \left(\frac{2}{\pi}\right)^{1/2}\frac{\mathrm{Ry}\,\tau_1}{\hbar}\left(\frac{x}{1+x}\right)^{1/2}\left(\frac{eA_{20}}{m_\mathrm{e}c^2}\right)^2\left(\frac{eA_{10}}{\hbar\omega}\right)^2$$

$$\times\frac{(\varepsilon_{0s}/I_0)^{3/2}}{(1+\varepsilon_{0s}/I_0)^4}\exp\left[-\left(\frac{\Delta\tau}{\tau_1}\right)^2\frac{2}{1+x}\right](1-2\cos^2\theta_0)^2. \quad (21)$$

The situation for $\tilde{t} = 1$ deserves special consideration, when $\varepsilon_{0s} \approx \hbar\omega$ (the production of the left satellite takes place at the threshold of ionisation) and the saddle point of the phase of the exponent in expression (18) for this satellite resides at the limit of the integration domain. In this case, the multiplier by the exponent in expression (18) should be expanded into a series about the point $\bar{z}_0 \approx z_0 - q \approx 0$. The expression for the intensity of the left satellite can be found with the aid of tabulated integrals [7] and has the form

$$\frac{\mathrm{d}\tilde{N}^{(\varepsilon_p=\varepsilon_{0s}-\hbar\omega)}(1)}{\mathrm{d}\Omega_{\boldsymbol{p}}} = \frac{4}{\pi}\left(\frac{\mathrm{Ry}\,\tau_1}{\hbar}\right)^{1/2}\left(\frac{x}{1+x}\right)^{1/4}$$

$$\times\left(\frac{eA_{20}}{m_\mathrm{e}c^2}\right)^2\left(\frac{eA_{10}}{\hbar\omega}\right)^2\left(\frac{\mathrm{Ry}}{I_0}\right)^{1/2}\frac{\varepsilon_{0s}/I_0}{(1+\varepsilon_{0s}/I_0)^4}$$

$$\times(1-2\cos^2\theta_0)^2\exp\left[-\left(\frac{\Delta\tau}{\tau_1}\right)^2\frac{2}{1+x}\right]. \quad (22)$$

The expression for the intensity of the right satellite is defined by expressions (19) and (20), in which one should take the upper sign and assume $\tilde{t} = 1$.

6. Analysis of results

We perform a comparative estimate of the intensities of the satellites and the main peak resulting from direct atom ionisation [expressions (16) and (17)]. The ratio between these quantities depends on the fundamental wave intensity, and the estimate will therefore give the value of required power at which the peak heights become equal. It follows from (16) and (17) for $\Delta\tau = 0$ and $\theta_0 = 0$ that

$$\frac{N^{(\varepsilon_p=\varepsilon_{0s}\pm\hbar\omega)}}{N^{(\varepsilon_p=\varepsilon_{0s})}} \sim \alpha^2\left(\frac{eA_{10}}{\hbar\omega}\right)^2. \quad (23)$$

For the 800-nm radiation of a Ti : sapphire laser, the ratio (23) achieves unity for the intensity $I_{10} \approx 2\times10^{12}$ W cm^{-2}, which corresponds to the experimental estimate of Ref. [3] obtained from the ponderomotive shift of photoelectron peaks.

We give the expression for the intensity ratio between the satellites produced due to Coulomb photoelectron rescattering from the ion [formula (21)] and those due to direct ionisation [formula (17)] in the case of high-lying peaks ($\tilde{t} = 0$):

$$\frac{\mathrm{d}\tilde{N}^{(\varepsilon_p=\varepsilon_{0s}\pm\hbar\omega)}(0)/\mathrm{d}\Omega_{\boldsymbol{p}}}{\mathrm{d}N^{(\varepsilon_p=\varepsilon_{0s}\pm\hbar\omega)}(0)/\mathrm{d}\Omega_{\boldsymbol{p}}} = \frac{1}{\varepsilon_{0s}/I_0}\frac{(1-2\cos^2\theta_0)^2}{\cos^4\theta_0}. \quad (24)$$

From expression (24) it follows that, with increase in main peak energy ε_{0s} (for instance, by going over to harmonics with higher numbers s), the relative contribution to the satellite production made by the Coulomb interaction of electrons in the continuum with the residual ion decreases, as would be expected. In particular, for He atoms ($I_0 =$ 24.6 eV) [3] this takes place for $s > s_0 = 2I_0/(\hbar\omega) \approx 31$.

Note that the inclusion of Coulomb interaction in this case significantly changes the angular dependence of photoelectrons in the satellites. In the case of direct ionisation, photoelectrons are primarily ejected along the wave polarisation direction ($\mathrm{d}N^{(\varepsilon_p=\varepsilon_{0s}\pm\hbar\omega)}/\mathrm{d}\Omega_{\boldsymbol{p}} \sim \cos^4\theta_0$) (this result was earlier obtained in Ref. [9]), while in the rescattering we have the expression

$$\frac{\mathrm{d}\tilde{N}^{(\varepsilon_p=\varepsilon_{0s}\pm\hbar\omega)}(0)}{\mathrm{d}\Omega_{\boldsymbol{p}}} \sim (1-2\cos^2\theta_0)^2.$$

It follows from expression (24), in particular, that the satellite formation in the direction $\theta_0 = \pi/2$ (i.e., in the transverse direction relative to the wave polarisation direction) takes place only due to inelastic Coulomb rescattering. We emphasise that this is true for satellites assuming that the ground state of the atom is the s state.

We give a comparative estimate of the heights of the satellite peaks arising from inelastic Coulomb rescattering [formula (19)] and direct photoionisation [formula (17)] in the general case, when $\tilde{t} \neq 0$ and $\tilde{t} \neq 1$:

$$\frac{\mathrm{d}\tilde{N}^{(\varepsilon_p=\varepsilon_{0s}\pm\hbar\omega)}/\mathrm{d}\Omega_{\boldsymbol{p}}}{\mathrm{d}N^{(\varepsilon_p=\varepsilon_{0s}\pm\hbar\omega)}/\mathrm{d}\Omega_{\boldsymbol{p}}} \approx \frac{1}{\varepsilon_{0s}/I_0}\frac{[\Phi^{(\pm)}(\tilde{t})]^2}{(1\pm\tilde{t})^5\cos^4\theta_0}. \quad (25)$$

In going over from the exact formula to approximate expression (25) we omitted the terms ε_{0s}/I_0 and $\tilde{t}$, which are small compared to unity. We employ the experimental parameters of Ref. [3] ($\lambda \approx 800$ nm, $s = 19$, $\varepsilon_{0s} = 4.7$eV, $I_0 = 24.6$ эВ, $\tilde{t} = 0.33$) to obtain from expression (25) the following expressions for the right satellite and the left one, respectively:

$$\frac{\mathrm{d}\tilde{N}^{(+)}/\mathrm{d}\Omega_{\boldsymbol{p}}}{\mathrm{d}N^{(+)}/\mathrm{d}\Omega_{\boldsymbol{p}}} \approx 2.0\frac{(1-3.0\cos^2\theta_0)^2}{\cos^4\theta_0}, \quad (26)$$

$$\frac{\mathrm{d}\tilde{N}^{(-)}/\mathrm{d}\Omega_{\boldsymbol{p}}}{\mathrm{d}N^{(-)}/\mathrm{d}\Omega_{\boldsymbol{p}}} \approx 15.1\frac{(1-0.83\cos^2\theta_0)^2}{\cos^4\theta_0}. \quad (27)$$

In the special case when $\theta_0 = 0$ (when photoelectrons are detected along the wave polarisation direction), the ratios (26) and (27) are equal to 8 and 0.4, respectively. We note that, according to expression (17), the right satellite always prevails over the left one in the case of direct photoionisation and the parameters adopted:

$$\frac{\mathrm{d}N^{(+)}/\mathrm{d}\Omega_{\boldsymbol{p}}}{\mathrm{d}N^{(-)}/\mathrm{d}\Omega_{\boldsymbol{p}}} \approx \frac{(1+\tilde{t})^{5/2}}{(1-\tilde{t})^{5/2}} \approx 5.$$

Naturally, the heights of the satellite peaks become equal when $\tilde{t} \to 0$. One can easily see that the abnormally small ratio (27) for the left satellite is related to the angular dependence of the satellite intensities.

To illustrate, for the parameters adopted we give the polar diagrams which define the angular dependences of the intensities of the satellites arising from inelastic Coulomb rescattering (Fig. 2). Of course, the asymmetry of the peaks noted in our work can be experimentally observed only when the high-order harmonic wave is a monochromatic wave with a definite number s.

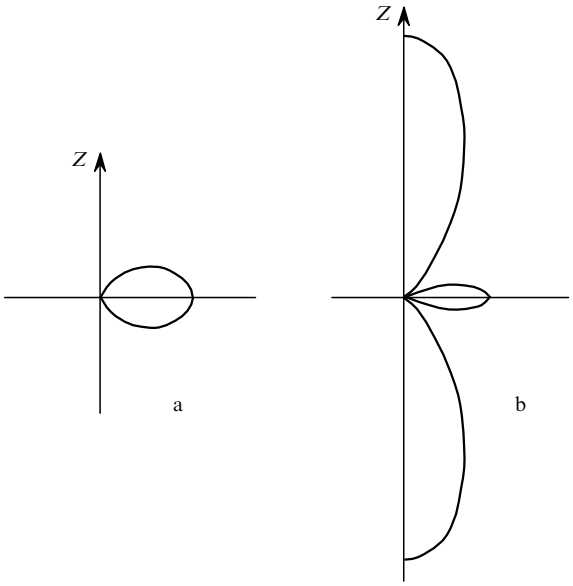

Figure 2. Polar diagrams describing the angular dependences of the intensities of the left (a) and right (b) satellites. The relative peak intensities are drawn to scale.

Note in conclusion that the results obtained in our work can provide a quantitative estimate of satellite intensities when the Born approximation is applicable for the photoelectron energies $\varepsilon_{0s} > I_0$. However, the qualitative results concerning the angular photoelectron redistribution in the satellites taking into account the Coulomb electron rescattering from ions are independent of electron energies. For this reason, the above quantitative estimates based on the experimental data [3] for $\varepsilon_{0s} \leqslant I_0$, are illustrative in character.

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Appendix 1

The substitution of (10) into (8) reduces the time integral to the expression

$$J_t = \int_{-\infty}^{\infty} \mathrm{d}t \left[1 + \Phi\left(\frac{t - t_0}{\tau_1\sqrt{x}}\right)\right] \exp\phi(t), \tag{A1.1}$$

where t_0 is the value of t at which the argument of the probability integral vanishes;

$$\phi(t) = \mathrm{i}[\varepsilon_p - (\varepsilon_{p'} \pm \omega)]t - \frac{(t - \Delta\tau)^2}{\tau_1^2} \tag{A1.2}$$

is the phase of the exponent.

The integral (A1.1) is calculated by the method of steepest descent and has the form [7]

$$J_t \approx \sqrt{\pi}\tau_1 \left[1 + \Phi\left(\frac{t_\mathrm{c} - t_0}{\tau_1\sqrt{x}}\right)\right] \exp\phi(t_\mathrm{c}), \tag{A1.3}$$

where $t_\mathrm{c} = \Delta\tau + \mathrm{i}[\varepsilon_\mathrm{p} - (\varepsilon_{p'} \pm \omega)]\tau_1^2/2$ is the coordinate of the saddle point obtained from the condition $\mathrm{d}\phi(t_\mathrm{c})/\mathrm{d}t = 0$.

Substitution of expression (A1.3) in expression (8) leads to the expression coincident with formula (11):

$$\begin{aligned}\tilde{A}^{(\varepsilon_p = \varepsilon_{0s} \pm \omega)} &= \frac{1}{\pi}(\mathrm{Ry}\,\tau_1)^2\sqrt{x}\,\frac{eA_{10}}{\omega}\frac{eA_{20}}{m_\mathrm{e}}(\pi a_0^3)^{1/2} \\ &\times \left(\frac{\mathrm{Ry}}{I_0}\right)^{3/4} \int_0^\infty \mathrm{d}z \frac{z}{(1 + \mathrm{Ry}\,z/I_0)^2}\left[1 + \Phi\left(\frac{t_\mathrm{c} - t_0}{\tau_1\sqrt{x}}\right)\right] \\ &\times \exp[-(z - z_0)^2\mathrm{Ry}^2\tau_1^2 x/4]\exp\{\mathrm{i}[\bar{z}_0 - (z \pm q)]\mathrm{Ry}\,\Delta t\} \\ &\times \exp\{-[\bar{z}_0 - (z \pm q)]^2\mathrm{Ry}^2\tau_1^2/4\} J_{\theta\varphi},\end{aligned} \tag{A1.4}$$

where we introduced dimensionless energies for convenience of calculation: $\bar{z}_0 = \varepsilon_p/\mathrm{Ry}$, $z = \varepsilon_{p'}/\mathrm{Ry}$, $z_0 = \varepsilon_{0s}/\mathrm{Ry}$ and $q = \omega/\mathrm{Ry}$. The integral

$$J_{\theta\varphi} = \int \frac{(\boldsymbol{e}\,\boldsymbol{e}_{p'})(\boldsymbol{e}\,\boldsymbol{e}_{\Delta p})}{[\bar{z}_0 + z - 2\sqrt{\bar{z}_0}z\cos(\boldsymbol{e}_p\,\boldsymbol{e}_{p'})]^{1/2}}\mathrm{d}\Omega_{p'} \tag{A1.5}$$

(where $\boldsymbol{e}_{\Delta p} = \Delta\boldsymbol{p}/|\boldsymbol{p} - \boldsymbol{p}'|$) is calculated over all possible angles of electron ejection in the intermediate state with a momentum $\boldsymbol{p}'$.

Appendix 2

We represent the integral in expression (14) in the general form as

$$J_z = \int_0^\infty f(z)\exp\phi(z)\mathrm{d}z, \tag{A2.1}$$

where $f(z)$ is the preexpotential multiplier (14) and the phase of the exponent is

$$\begin{aligned}\phi(z) &= \mathrm{i}[\bar{z}_0 - (z \pm q)]\mathrm{Ry}\,\Delta t - \frac{(z - z_0)^2\mathrm{Ry}^2\tau_1^2 x}{4} \\ &- \frac{[\bar{z}_0 - (z \pm q)]^2\mathrm{Ry}^2\tau_1^2}{4}.\end{aligned} \tag{A2.2}$$

The integral (A2.1) is calculated by the method of steepest descent [7]; the coordinate of the saddle point is

$$z_\mathrm{c} = \frac{z_0 x + \bar{z}_0 \mp q}{1 + x} - \mathrm{i}\frac{2\Delta\tau}{\mathrm{Ry}\,\tau_1^2(x + 1)} = z_0 + \mathrm{O}\left(\frac{1}{\omega\tau_1}\right). \tag{A2.3}$$

By integrating (A2.1), we obtain the expression

$$J_z = \frac{4\sqrt{\pi}}{\mathrm{Ry}\,\tau_1(1+x)^{1/2}} \frac{F_0(\bar{z}_0) + F_1(\bar{z}_0)\cos^2\theta_0}{(1+\mathrm{Ry}\,z_0/I_0)^2\bar{z}_0^{3/2}}$$
$$\times \exp\left\{ -\frac{[\bar{z}_0 - (z \pm q)]^2 \mathrm{Ry}^2\tau_1^2 x}{4(1+x)} \right\}$$
$$\times \exp\left[-\left(\frac{\Delta\tau}{\tau_1}\right)\frac{1}{1+x} \right], \qquad \text{(A2.4)}$$

in which the argument z of the functions $F_0(z)$ and $F_1(z)$ (13) was taken for $z = \bar{z}_0$.

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