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Modification of the spectrum of high harmonics by a weak vacuum ultraviolet field

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Abstract. **Within the framework of the time-dependent effective range theory, we have calculated the spectra of high harmonics in a field containing an intense low-frequency component and a weak (perturbative) high-frequency component with a photon energy exceeding the ionisation potential of the atomic target. We have presented a quantum-mechanical substantiation of the quasi-classical theory [A.V. Flegel et al. *Quantum Electron.*, 47, 222 (2017)], which describes additional generation channels of high harmonics induced by a weak high-frequency field.**

Keywords: *high harmonic generation, effective range theory, strong field, vacuum ultraviolet.*

Investigations of high harmonic generation (HHG) are closely related to a wide range of practical applications, in particular, the generation of extremely short laser pulses [1–3] and the development of new methods for spectroscopy of ultrafast processes based on the analysis of HHG spectra [4–8]. A characteristic feature of HHG in strong laser fields is the appearance of a plateau in the spectrum of high harmonics, i.e., a weak dependence of the harmonic yield on its energy E_Ω. The number and shape of plateau-like structures in the HHG spectrum depend substantially on the laser pulse structure. For example, in the simplest case of a monochromatic linearly polarised field, one plateau is observed with a cut-off energy $E_c \approx |E_0| + 3.17u_p$, where E_0 is the binding energy of the external electron; $u_p = F^2/(4\omega^2)$ is the average vibrational energy of an electron in a field of intensity F and frequency ω [9] (hereafter the atomic system of units is used). In this case, the height of the plateau and the cut-off position can be controlled only by changing F and ω. The addition of electric or magnetic fields to the initial intense field leads to a modification of the plateau-like structure in the HHG spectrum, as well as to an increase in the number of parameters that allow us to control the shape of the high harmonic spectrum.

Recently, a two-component HHG scheme has attracted a lot of attention, in which a high-frequency component with one or several carrier frequencies lying in the UV range is added to the intense IR field [10–12]. Adding a weak UV component leads to a significant modification of the spectrum of high harmonics. If the energy of the UV photon does not exceed the ionisation potential of the atomic system, it is possible to increase the efficiency of the yield of high harmonics due to resonant population of the excited states of the system [10, 11, 13, 14]. The use of high frequency fields lying in the vacuum UV range leads to the appearance of additional plateau-like structures of both one-electron [15–17] and many-electron [18–20] origin. Flegel et al. [17] proposed to use the quasi-classical analysis of the dynamics of an electron in a two-component field to solve the problem under consideration and estimated the cut-off position of the emerging plateau-like structures.

The purpose of this paper is to calculate quantum-mechanically the high harmonic spectrum in the framework of the effective range theory [21, 22] and to determine the accuracy of the quasi-classical estimates of the position of spectral cut-off of plateau structures obtained in Ref. [17].

Consider an atomic system with a valence *s*-electron with a binding energy $E_0 = -\kappa^2/2$ in a two-component laser field. We parametrize a two-component laser field with a frequency ω and a strength F, determining the IR component, and also with a frequency $\Omega = k\omega$ (k is an odd integer) and an intensity F_k for the UV component of the field,

$$\boldsymbol{F}(t) = \boldsymbol{e}_z[F\cos(\omega t) + F_k\cos(\Omega t + \phi)], \tag{1}$$

where ϕ is the phase shift. Note that the choice of the spatial orientation of the field components does not change the structure of the high-energy part of the high harmonic spectrum, but leads to a modification of the polarisation properties of the harmonics.

The interaction of the atomic system with a time-periodic field of form (1) can be described in the framework of the model approach combining the time-dependent efective range theory [21, 22] and the method of complex quasi-energy [23]. Within the framework of this approach, the wave function of the quasi-stationary quasi-energy state of the atomic system in an external periodic field can be given in the form [24]

$$\begin{gathered}\Phi_\varepsilon(\boldsymbol{r},t) = -C_0\sqrt{\frac{\kappa}{\pi}}\sum_q f_q\Phi_\varepsilon^{(q)}(\boldsymbol{r},t),\\ \Phi_\varepsilon^{(q)}(\boldsymbol{r},t) = \int_{-\infty}^{t}\exp[\mathrm{i}\varepsilon(t-t')]G^{(+)}(\boldsymbol{r},t;0,t')\exp(-2\mathrm{i}q\omega t')\,\mathrm{d}t',\end{gathered} \tag{2}$$

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where C_0 is the dimensionless asymptotic coefficient of the unperturbed wave function of the bound state; $G^{(+)}(\boldsymbol{r},t;0,t')$ is the retarded Green's function in the laser field;

$$G^{(+)}(\boldsymbol{r},t;\boldsymbol{r}',t') = G_0^{(+)}(\boldsymbol{r},t;\boldsymbol{r}',t')\exp[\mathrm{i}R(\boldsymbol{r},t;\boldsymbol{r}',t') - \mathrm{i}S(t,t')];$$

$$G_0^{(+)}(\boldsymbol{r},t;\boldsymbol{r}',t') = -\mathrm{i}\left[\frac{1}{2\pi\mathrm{i}(t-t')}\right]^{3/2}\exp\left[\mathrm{i}\frac{(\boldsymbol{r}-\boldsymbol{r}')^2}{2(t-t')}\right];$$

$$R(\boldsymbol{r},t;\boldsymbol{r}',t') = \boldsymbol{r}\boldsymbol{P}(t;t,t') - \boldsymbol{r}'\boldsymbol{P}(t';t,t'); \tag{3}$$

$$S(t,t') = \frac{1}{2}\int_{t'}^{t}\boldsymbol{P}(\tau;t,t')^2\mathrm{d}\tau;$$

$$\boldsymbol{P}(\tau;t,t') = \frac{1}{c}\left(\boldsymbol{A}(\tau) - \frac{1}{t-t'}\int_{t'}^{t}\boldsymbol{A}(\tau)\mathrm{d}\tau\right);$$

$\boldsymbol{A}(\tau)$ is the vector potential of the two-component field; and c is the speed of light in the vacuum. The coefficients f_q are found from the system of homogeneous equations for the complex quasi-energy ε,

$$\sum_{q'}\mu_{qq'}(\varepsilon)f_{q'} = 0, \tag{4}$$

where the matrix element $\mu_{qq'}(\varepsilon)$ can be expressed as a double integral:

$$\mu_{qq'}(\varepsilon) = \mathcal{R}_0(\varepsilon+2q\omega)\delta_{qq'} - M_{qq'}(\varepsilon); \tag{5}$$

$$M_{qq'}(\varepsilon) = \sqrt{\frac{1}{2\pi\mathrm{i}}}\frac{1}{T}$$
$$\times\int_0^T\mathrm{d}t\int_{-\infty}^{t}\frac{\mathrm{d}t'\exp\{\mathrm{i}[\varepsilon(t-t') + 2q\omega t - 2q'\omega t']\}}{(t-t')^{3/2}}$$
$$\times\{\exp[-\mathrm{i}S(t,t')] - 1\}. \tag{6}$$

The function $\mathcal{R}_0(E)$ can be expressed in terms of the main parameters of the effective range theory, i.e. the scattering length a_0 and the effective range r_0:

$$\mathcal{R}_0(E) = -\frac{1}{a_0} + r_0E - \mathrm{i}\sqrt{2E}. \tag{7}$$

The solution of system (4) makes it possible to find the shift and level broadening in an external laser field. However, as analysis shows, the allowance for the above correction in the HHG problem makes a negligible contribution to the amplitude of the process and the exact value of quasi-energy can be replaced by the binding energy of the valence electron in the absence of the field, $\varepsilon \approx E_0$. Then, to find the coefficients f_q, system (4) can be solved by using the iterative procedure [25]. After choosing $f_q = \delta_{q0}$ as a zeroth approximation, we obtain the following expressions for the coefficients $f_{q\neq 0}$:

$$f_{q\neq 0} \approx \frac{M_{q0}}{\mathcal{R}_0(E_0+2q\omega)}. \tag{8}$$

Knowing the wave function (2), we can write the HHG amplitude with a given polarisation $\boldsymbol{e}_h$ in the form [24]:

$$A_N = (\boldsymbol{e}_h^*\boldsymbol{d}_N), \tag{9}$$

$$\boldsymbol{d}_N = \frac{1}{T}\int_0^T\mathrm{d}t\mathrm{e}^{\mathrm{i}N\omega t}\langle\tilde{\Phi}_\varepsilon(\boldsymbol{r},t)\,|\,\boldsymbol{r}\,|\,\Phi_\varepsilon(\boldsymbol{r},t)\rangle, \tag{10}$$

where $\tilde{\Phi}_\varepsilon(\boldsymbol{r},t)$ is the dual function obtained from the quasi-stationary quasi-energy state function $\Phi_\varepsilon(\boldsymbol{r},t)$ by complex conjugation and time reversal [24, 26]. Taking into account expansion (2), the Fourier transform of the dipole moment $\boldsymbol{d}_N$ can be written in the form of a double sum containing the coefficients f_q:

$$\boldsymbol{d}_N = \sum_{qq'}f_{q'}\boldsymbol{D}_{qq'}f_{q'}, \tag{11}$$

$$\boldsymbol{D}_{qq'}(N) = -\sqrt{\frac{1}{2\pi\mathrm{i}}}\frac{\kappa C_0^2}{N\omega T}$$
$$\times\int_0^T\mathrm{d}t\int_{-\infty}^{t}\frac{\mathrm{d}t'\exp[\mathrm{i}\varepsilon(t-t') + 2\mathrm{i}q\omega t]}{(t-t')^{3/2}}$$
$$\times\exp[-2\mathrm{i}q'\omega t' - \mathrm{i}S(t,t')]\int_{t'}^{t}\boldsymbol{P}(\tau;t,t')\exp(\mathrm{i}N\omega\tau)\mathrm{d}\tau. \tag{12}$$

If the UV pulse parameters satisfy the condition $\kappa\Omega/F_k \gg 1$, the interaction with this field can be considered in the framework of perturbation theory [27]. In this case, the HHG amplitude in a two-component laser field can be represented in the form of partial amplitudes describing HHG with the participation of n-photons of the UV field:

$$\boldsymbol{d}_N = \boldsymbol{e}_z\sum_n d_N^{(n)}. \tag{13}$$

We present the first two terms of this expansion in the form:

$$d_N^{(0)} = \sum_{qq'}f_q^{(0)}D_{qq'}^{(0)}f_{q'}^{(0)}, \tag{14}$$

$$d_N^{(1)} = \sum_{qq'}(f_q^{(0)}D_{qq'}^{(1)}f_{q'}^{(0)} + f_q^{(1)}D_{qq'}^{(0)}f_{q'}^{(0)} + f_q^{(0)}D_{qq'}^{(0)}f_{q'}^{(1)}). \tag{15}$$

To analyse the relative contribution of processes occurring with the exchange of n-photons of the UV field, it is also convenient to introduce the partial probabilities of the HHG process:

$$R_N^{(n)} = \frac{(N\omega)^3}{2\pi c^3}|d_N^{(n)}|^2. \tag{16}$$

In the case when the IR and UV components of the field are monochromatic, the expressions for the matrix elements $D_{qq'}^{(i)}$ and $M_{0q}^{(i)}$ [determine the coefficients $f_q^{(i)}$ in accordance with (8)] can be represented as one-dimensional integrals of the Bessel function. For example, for the zero-order matrix elements they have the form [22]

$$M_{qq'}^{(0)} = \mathrm{i}^{q-q'}\sqrt{\frac{\omega}{2\pi\mathrm{i}}}\int_0^\infty\frac{\exp[\mathrm{i}(E_0/\omega + q + q')\tau]}{\tau^{3/2}}$$

$$\times[\exp(-\mathrm{i}\lambda(\tau))J_{q-q'}(z(\tau))-\delta_{qq'}]\mathrm{d}\tau, \quad (17)$$

$$D^{(0)}_{qq'}(n)=\mathcal{N}\int_0^{\infty}\frac{\mathrm{d}\tau}{\tau^{3/2}}\exp[\mathrm{i}(E_0/\omega+q+q')\tau-\mathrm{i}\lambda(\tau)]$$

$$\times[j_-(\tau)\mathcal{J}_{-1}(\tau)-\mathrm{i}j_+(\tau)\mathcal{J}_{+1}(\tau)], \quad (18)$$

where

$$\mathcal{J}_n(\tau)=J_\eta(z(\tau));\quad \eta=q-q'+(N+n)/2;$$

$$z(\tau)=\frac{2u_\mathrm{p}}{\omega}\sin\frac{\tau}{2}\left[\cos\frac{\tau}{2}-2\frac{\sin(\tau/2)}{\tau}\right];$$

$$\lambda(\tau)=\frac{u_\mathrm{p}}{\omega}\left[\tau-4\frac{\sin^2(\tau/2)}{\tau}\right];$$

$$j_\pm(\tau)=2\frac{\sin(\tau/2)\sin(N\tau/2)}{\tau}-\frac{N\sin[(N\pm1)\tau/2]}{N\pm1};$$

$$\mathcal{N}=-\mathrm{i}^{q-q'+N/2}\frac{C_0^2\kappa}{N^2}\sqrt{\frac{u_\mathrm{p}}{\pi\omega}}.$$

For first-order corrections with respect to the strength F_k, the corresponding matrix elements have the form:

$$M^{(1)}_{qq'}=-\frac{F_k}{\kappa C_0^2}[D^{(0)}_{qq'}(k)\mathrm{e}^{\mathrm{i}\phi}+D^{(0)}_{qq'}(-k)\mathrm{e}^{-\mathrm{i}\phi}],$$

$$D^{(1)}_{qq'}(N)=\sum_{\nu,\mu=\pm1}\mathrm{e}^{\mathrm{i}\mu\phi}D^{(\mu,\nu)}_{qq'}(N,k), \quad (19)$$

where

$$D^{(\mu,\nu)}_{qq'}(N,k)=-\mathcal{N}\mathrm{i}^{(1+\mu k)/2}\frac{F_k}{\mu k}$$

$$\times\int_0^{\infty}\frac{\exp[\mathrm{i}(E_0/\omega+q+q')\tau-\mathrm{i}\lambda(\tau)]}{\tau^{3/2}}\exp(\mathrm{i}k\nu\tau/2)$$

$$\times\Big\{w^{\mu,\nu}_{N,k}(\tau)\mathcal{J}_{\mu k}(\tau)-2\frac{u_\mathrm{p}}{\omega}[w^{-\mu,\nu}_{1,k}(\tau)j_-(\tau)\mathcal{J}_{\mu k-2}(\tau) \quad (20)$$

$$-w^{\mu,\nu}_{1,k}(\tau)j_+(\tau)\mathcal{J}_{\mu k+2}(\tau)-\mathrm{i}(w^{\mu,\nu}_{1,k}(\tau)j_-(\tau)$$

$$+w^{-\mu,\nu}_{1,k}(\tau)j_+(\tau))\mathcal{J}_{\mu k}(\tau)]\Big\}\mathrm{d}\tau;$$

$$w^{\mu,\nu}_{n,k}=\frac{\nu}{2\mathrm{i}}\left[\frac{2\sin(n\tau/2)}{nk\tau}-\mu\frac{\exp(\mathrm{i}\mu\nu n\tau/2)}{n+\mu k}\right]. \quad (21)$$

To analyse the influence of the UV field on the HHG process, we calculated the HHG spectra for an atomic system with a binding energy E_0 = –13.605 eV and an asymptotic coefficient C_0 = 2 (corresponding to the parameters of the hydrogen atom). The frequency of the IR component corresponds to a wavelength λ = 1200 nm (ω = 0.038 at. units), and $\Omega = 41\omega$ or 51ω. The intensity of the main component is $I = 2\times10^{14}$ W cm^{-2}, and the intensity of the additional field is 1% of the intensity of the main field. Note that in the first order of perturbation theory the partial yield of $R_N^{(1)}$ depends linearly on the intensity of the UV component of the field. At the same time, as seen from (20) and (21), the dependence of the partial yield on the frequency of the UV photon cannot be expressed as an elementary function. The partial probabilities of $R_N^{(0)}$ and $R_N^{(1)}$ are shown in Fig. 1. For the partial yield of $R_N^{(1)}$, two plateau structures with different lengths and intensities are observed. The cut-off energy of a shorter plateau is smaller than that for a monochromatic IR field. When the energy of the UV photon is increased, the cut-off energy of the short plateau decreases. The cut-off energy of a long plateau exceeds the maximum energy in a monochromatic IR field by exactly the energy of the UV photon.

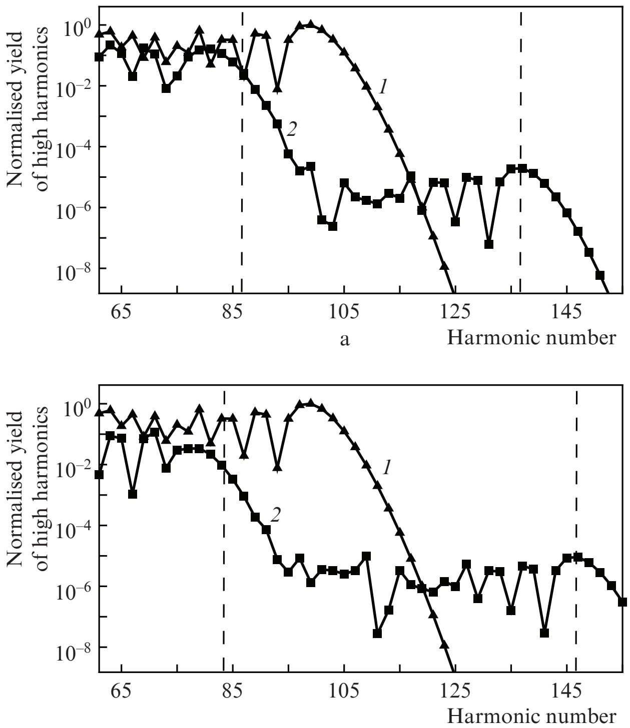

Figure 1. Dependences of the partial yields of high harmonics $R_N^{(0)}$ (*1*) and $R_N^{(1)}$ (*2*) on the harmonic number for an atomic system with binding energy E_0 = –13.605 eV in a two-component laser field. The wavelength of the IR component is λ = 1200 nm, and the intensity is $I = 2\times10^{14}$ W cm^{-2}. The wavelength of the UV component with $I_k = 2\times10^{12}$ W cm^{-2} corresponds to the (a) 41st and (b) 51st harmonics of the IR component. The results are normalised to the condition $R_{99}^{(0)} = 1$, where N = 99 corresponds to the plateau cut-off position in a monochromatic IR field. Here and in Fig. 2, the vertical dashed lines indicate the position of the plateau cut-off in accordance with the quasi-classical estimates proposed in [17].

This behaviour of the cut-off position of plateau-like structures is in agreement with the quasi-classical theory proposed in [17]. In accordance with it, the emergence of an intense short plateau is associated with the absorption of the UV photon during the ionisation stage, whereas the less intense long plateau arises from the absorption of the UV photon during the recombination stage. In [17], analytical expressions for estimating the boundaries of high-energy plateaus are also given. The cut-off positions of the plateau-like structures calculated in accordance with the indicated analytical expressions are shown in Fig. 1 by vertical dashed lines. One can see that the quasi-classical theory describes with good accuracy the position of the cut-off of a long plateau, but for a short plateau the estimates of the cut-off position given by it lead to overestimated results. The good agreement of the cut-off position of the long plateau with the classical estimates proposed in [17] is due to the fact that the dynamics of the electron in the continuum is determined by the zero initial velocity and, hence, by the minimal spreading of the wave packet formed as a result of electron tunnelling from the bound state. In the case of a short plateau, the transition of an electron to a continuum occurs through the absorption of a UV photon, which corresponds to a nonzero initial electron velocity in the continuum. The spreading of such a packet occurs much faster; as a result, the actual cut-off position of the plateau differs significantly from the classical result.

An important approximation, widely used for calculations in strong laser fields, is the strong field approximation (SFA). It consists in neglecting the effects of the atomic potential at the stage of electron motion in the field-modified continuum. Within the framework of the effective range theory, SFA is reduced to the substitution $f_q = \delta_{q,0}$, and the dipole matrix element $\boldsymbol{d}_N$ takes the form:

$$\boldsymbol{d}_N = \boldsymbol{e}_z \sum_{\nu,\mu=\pm 1} \mathrm{e}^{\mathrm{i}\mu\varphi} D_{0,0}^{(\mu,\nu)}(N,k). \tag{22}$$

Moreover, each of the terms in sum (22) has a transparent physical meaning. Obviously, the terms with $\mu = +1$ correspond to the emission of the UV photon in the HHG process, and the terms with $\mu = -1$ correspond to the absorption of the UV photon. To understand the meaning of the variable ν, it is necessary to turn to the general expression for the dipole moment (11). It is seen that the expression for any matrix element is represented as a double integral with respect to the variables t and t', which can be interpreted as the starting and ending times of motion in the field-modified continuum. The value of the variable ν corresponds to the instant of time in which the interaction with the UV field occurs: for $\mu = \nu$ this is the moment of ionisation (t'), and for $\mu = -\nu$ this is the moment of return of the electron to the parent core (t). It should be noted that, in addition to the cases described above, the exchange of a UV photon is also possible at the stage of electron motion in the continuum. The HHG amplitude in the corresponding channel is described by terms containing nonzero coefficients $f_q^{(1)}$. Thus, the analysis of the accuracy of the SFA also makes it possible to estimate the relative contribution of the HHG channel to the emission or absorption of the UV photon at the stage of electron motion in the continuum.

Figure 2 shows the partial probabilities

$$R_N^{(\mu,\nu)} = \frac{(N\omega)^3}{2\pi c^3} |D_{00}^{(\mu,\nu)}(N,k)|^2, \tag{23}$$

corresponding to the absorption of the UV photon in the stage of ionisation ($R_N^{(-,-)}$) and recombination ($R_N^{(-,+)}$), calculated for a two-component field with the same parameters as in Fig. 1. It can be seen that the contributions of the partial yields $R_N^{(-,-)}$ and $R_N^{(-,+)}$ completely describe the observed plateau-like structures in the HHG spectrum. This indicates that in the first order of perturbation theory in terms of the intensity of the UV pulse, only two channels considered in [17] make the main contribution to the total yield of HHG, and the contribution of the remaining channels is negligibly small.

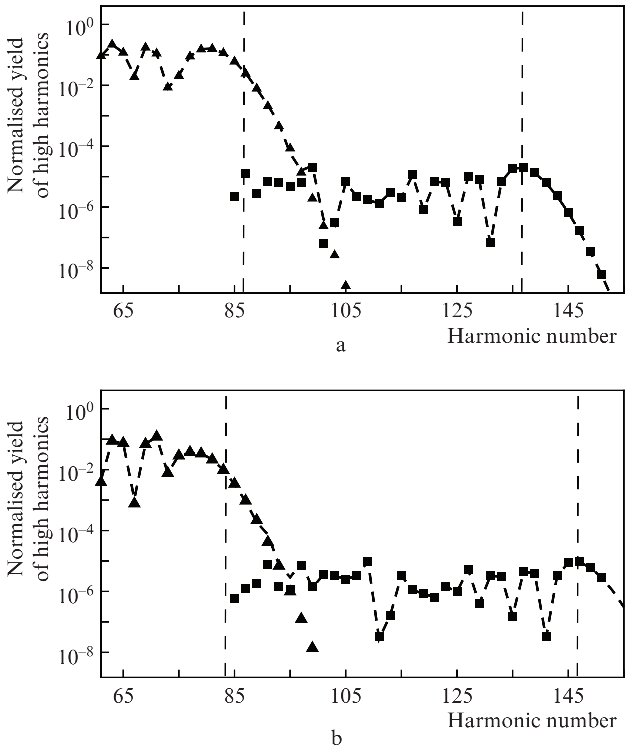

Figure 2. Dependences of the partial yields of high harmonics $R_N^{(1)}$ (dashed curves), $R_N^{(-,-)}$ (triangles) and $R_N^{(-,+)}$ (squares) on the harmonic number in a two-component laser field. The calculation parameters are the same as in Fig. 1.

In conclusion, let us formulate the main results of this paper. Using the time-dependent effective range theory, for a two-component field consisting of a low-frequency (IR) and a high-frequency (UV) components we obtained analytical expressions for the HHG amplitude in the first order of perturbation theory in terms of the strength of the UV field both in the strong-field approximation and with the effects of rescattering of higher orders taken into account. We have shown that in the strong-field approximation the analytic expression for the HHG amplitude allows one to single out the contributions of the channels corresponding to the emission/absorption of the UV photon during the ionisation or recombination stage. It follows from the calculations that in the first order of perturbation theory in terms of the intensity of the UV field, the emerging plateau-like structures are completely described by the contribution from the channels corresponding to the absorption of the UV photon during the stages of ionisation and recombination. Thus, the quantum-mechanical calculations carried out qualitatively confirm the conclusions of paper [17].

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