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Tunnelling approximation for estimating the amplitude of high harmonic generation in intense laser fields: analysis of ionisation and recombination times

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Abstract. **We consider in the low-frequency approximation the quasi-classical equations for the ionisation and recombination times that arise in the analysis of high harmonic generation in the quasi-classical approximation. Based on the assumption that the ratio of the time of the subbarrier motion of an atomic electron, $\Delta t'$, to the characteristic period of the laser pulse, T, is small, we have constructed a formal perturbation theory with respect to the parameter $\Delta t'/T$ to calculate the ionisation and recombination times. The accuracy of the constructed perturbation theory is analysed. Using the approach developed, we suggest extending the classical theory of rescattering for linear polarisation to the case of fields with two spatial components.**

Keywords: *high harmonic generation, quasi-classical approximation, strong laser field, analytical theories.*

1. Introduction

One of the most commonly used analytical methods for analysing nonlinear phenomena induced by a strong laser field is the S-matrix approach [1–4]. In the framework of this approach, a strong laser field is taken into account exactly, while a series of perturbation theory is formally constructed from the atomic (or molecular) potential. As a result of such an expansion, the amplitude of the process induced by a strong laser field can be represented in the form of integrals of rapidly oscillating functions. For example, in the case of high harmonic generation (HHG), it can be written in the form*:

$$A = \int_{-\infty}^{\infty} \mathrm{d}t \int_{-\infty}^{t} \mathrm{d}t' f(t,t') \exp[\mathrm{i}S_{\mathrm{tot}}(t,t')], \tag{1}$$

$$S_{\mathrm{tot}}(t,t') = S_{\mathrm{cl}}(t,t') + \Omega t, \tag{2}$$

$$S_{\mathrm{cl}}(t,t') = -\frac{1}{2}\int_{t'}^{t}\left[\boldsymbol{A}(\xi) - \frac{1}{t-t'}\int_{t'}^{t}\boldsymbol{A}(\xi')\mathrm{d}\xi'\right]^2 \mathrm{d}\xi - I_{\mathrm{p}}(t-t'), \tag{3}$$

where $f(t,t')$ is a smooth function; $S_{\mathrm{cl}}(t,t')$ is the classical action of a particle in a laser field; $\boldsymbol{A}(t)$ is the vector potential of the laser field; Ω is the frequency of the emitted photon; and I_{p} is the ionisation potential of the atomic target. If the carrier frequency of the laser pulse is much smaller than the ionisation potential I_{p}, then the two-dimensional integral (1) can be estimated with good accuracy by the saddle-point method [1–3, 5], and the HHG amplitude can be presented as the sum of the partial amplitudes A_j:

$$A = \sum_j A_j. \tag{4}$$

Each partial amplitude A_j is associated with the jth closed trajectory (quantum orbit), determined by the times of the beginning of the motion (t_j') and the return to the initial point (t_j) [1, 5]. From the mathematical point of view, the times t_j' and t_j are the saddle points of integrand (1), and the corresponding system of equations for t_j' and t_j is determined by zeroing the first derivatives with respect to t and t' from the phase function of integrand (1):

$$\boldsymbol{K}'^2(t,t')/2 = -I_{\mathrm{p}}, \tag{5a}$$

$$\boldsymbol{K}^2(t,t')/2 = E, \tag{5b}$$

where

$$\boldsymbol{K}'(t,t') = \boldsymbol{A}(t') - \frac{1}{t-t'}\int_{t'}^{t}\boldsymbol{A}(\xi)\mathrm{d}\xi, \tag{6a}$$

$$\boldsymbol{K}(t,t') = \boldsymbol{A}(t) - \frac{1}{t-t'}\int_{t'}^{t}\boldsymbol{A}(\xi)\mathrm{d}\xi \tag{6b}$$

and $E = \Omega - I_{\mathrm{p}}$.

From the physical point of view, the system of equations (5) determines the quasi-classical conditions for realising the transition from the bound state to the state of the continuous spectrum (5a) and from the state of the continuous spectrum to the bound state with simultaneous emission of a photon with energy Ω (5b). Indeed, in the quasi-classical approximation, transitions occur at the time moment (in the general case, complex), for which the energies of the initial and final states are equal [6].

An elementary analysis of the system of equations (5) shows that the solution of the system is possible only on the set of complex numbers, and consequently the closed trajectory corresponding to the pair of times $\{t_j', t_j\}$ is also complex. In most cases, for a physical interpretation of features in the

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HHG spectra, it is necessary to associate a classical (real) trajectory with a complex trajectory, which is found from the classical limit of system (5). As a rule, the classical limit of the system of equations (5) is determined by the formal limit $I_p \to 0$. In the case of linear polarisation of the field, this means that an electron is released from an atom with a zero initial energy, propagates in a laser field along a closed classical trajectory and, after emitting a photon with energy Ω, forms a bound state. It is obvious that the system of equations (5) in its classical limit provides a theoretical justification for the classical rescattering model [7, 8]. In contrast, in the case of laser fields with two spatial components of the field with different projections of the electric vector on the x and y axes, the limit $I_p \to 0$ does not allow one to find a closed classical trajectory, in which case the interpretation of the HHG spectra in two-component fields is based on the formalism of missed trajectories. Within the framework of this formalism, it is asserted that the absence of closed trajectories with a zero initial energy causes an effective suppression of the electron recombination process in HHG [9–13]. A significant drawback of this interpretation is that recombination is a purely quantum process, and its probability depends only on the properties of the atomic target.

In the present paper it is shown that the system of equations (5) admits limiting solutions to which there correspond closed classical (real) trajectories in a two-component laser field. Thus, the interpretation of the HHG spectra in a two-component laser field (as in the case of a linear field) can be performed within the framework of the concept of closed trajectories.

2. Analysis of the HHG amplitude in the tunnelling limit

Let us estimate expression (1) by successively calculating the integrals in t' and t by the saddle-point method. It is well known that in the quasi-classical approximation the major contribution is made only by those trajectories that are determined by the times with minimal imaginary parts [1, 14]. To find such trajectories, we use explicitly the real and imaginary parts in $t' = \bar{t}' + i\Delta t'$ and expand equation (5a), which determines the saddle points with respect to t' in a series with respect to $\Delta t'$ to the third order:

$$\boldsymbol{K}'^2 + \kappa^2 + i\Delta t' \dot{\boldsymbol{K}}'^2 - \frac{\Delta t'^2}{2}\ddot{\boldsymbol{K}}'^2 - i\frac{\Delta t'^3}{6}\dddot{\boldsymbol{K}}'^2 = 0, \tag{7}$$

where

$$\kappa^2 = 2I_p; \quad \boldsymbol{K}' \equiv \boldsymbol{K}'(t, \bar{t}'); \quad \dot{\boldsymbol{K}}'^2 \equiv \frac{\partial \boldsymbol{K}'^2}{\partial \bar{t}'};$$

$$\ddot{\boldsymbol{K}}'^2 \equiv \frac{\partial^2 \boldsymbol{K}'^2}{\partial \bar{t}'^2}; \quad \dddot{\boldsymbol{K}}'^2 \equiv \frac{\partial^3 \boldsymbol{K}'^2}{\partial \bar{t}'^3}.$$

Note that the time of the subbarrier motion of an electron in a low-frequency field, determined by the imaginary part of t', can be considerably smaller than the characteristic period of the laser pulse, $T = 2\pi/\omega$, where ω is the carrier frequency [15–18]. Taking into account that the characteristic order of the nth derivative in (7) is proportional to $\boldsymbol{K}'^2/T^n$, one can easily see that each successive term in the expansion in (7) with respect to the preceding one is on the order of smallness $\Delta t'/T$. Thus, the range of applicability of expansion (7) is determined by the ratio $\Delta t'/T$. Selecting the imaginary and real parts in equation (7), we obtain two equations:

$$\boldsymbol{K}'^2 + \kappa^2 - \frac{\Delta t'^2}{2}\ddot{\boldsymbol{K}}'^2 = 0, \tag{8a}$$

$$\dot{\boldsymbol{K}}'^2 - \frac{\Delta t'^2}{6}\dddot{\boldsymbol{K}}'^2 = 0. \tag{8b}$$

Expressing $\Delta t'$ from equation (8a) as

$$\Delta t' = \sqrt{\frac{2(\kappa^2 + \boldsymbol{K}'^2)}{\ddot{\boldsymbol{K}}'^2}} \tag{9}$$

and substituting it in (8b), we obtain

$$\dot{\boldsymbol{K}}'^2 - \frac{1}{3}\frac{(\kappa^2 + \boldsymbol{K}'^2)}{\ddot{\boldsymbol{K}}'^2}\dddot{\boldsymbol{K}}'^2 = 0. \tag{10}$$

The solution of equation (10) defines the initial (real) time of motion along a closed trajectory and also determines the time of subbarrier motion [see relation (9)]. The second term in (10) is determined by the time of subbarrier motion and has a quantum nature; therefore, we will call this term the quantum correction to the condition on the energy extremum at the initial instant $\bar{t}'$.

Let us study qualitatively equation (10). The second term as a correction to the first term in the zeroth approximation can be omitted. Then, equation (10) is transformed to the form

$$\dot{\boldsymbol{K}}'^2 = 0. \tag{11}$$

Equation (11) has a transparent physical meaning. Namely, it determines the extremum (minimum) points of the electron energy at the initial moment of motion along a closed trajectory. We note that at the minimum point the second derivative is positive ($\ddot{\boldsymbol{K}}'^2 > 0$), which obviously provides a positive value of the radicand in (9). Thus, the electron begins its motion along a closed trajectory at a time when its energy in the laser field is minimal. Note that in the case of linear polarisation, condition (11) is equivalent to $K' = 0$.

When integrating (1) with respect to t' by the saddle-point method, it is necessary to find the second derivative of the phase function at the saddle point:

$$\alpha(t) = \frac{\partial^2 S_{cl}(t,t')}{\partial t'^2} = \frac{1}{2}\frac{\partial \boldsymbol{K}'^2(t,t')}{\partial t'} \approx \frac{i}{2}\Delta t' \ddot{\boldsymbol{K}}'^2, \tag{12}$$

and also to separate the real and imaginary parts in the phase function. Expanding S_{cl} (3) in a series in $\Delta t'$ to the third summand, we obtain

$$S_{cl}(t,t') \approx S_{cl}(t,\bar{t}') + i\frac{\Delta t'}{2}(\boldsymbol{K}'^2 + \kappa^2) - \frac{(\Delta t')^2}{4}\dot{\boldsymbol{K}}'^2 - i\frac{(\Delta t')^3}{12}\ddot{\boldsymbol{K}}'^2$$

$$= S_{cl}(t,\bar{t}') + \frac{i}{3}\sqrt{\frac{2(\kappa^2 + \boldsymbol{K}'^2)}{\ddot{\boldsymbol{K}}'^2}}(\kappa^2 + \boldsymbol{K}'^2). \tag{13}$$

Note that, in deriving (13), we must take into account relation (11) rather than (10), since the allowance for the latter leads to an excess of the accuracy of approximate calculations. As a result of calculating the integral in t' we obtain

$$A = \sqrt{2\pi}\sum\int_{-\infty}^{\infty} g(t)\exp[\mathrm{i}S_{\mathrm{cl}}(t,\bar{t}') + \mathrm{i}\Omega t]\,\mathrm{d}t, \tag{14}$$

where

$$g(t) = \frac{f(t,\bar{t}')\exp[-\Lambda^3(t,\bar{t}')/3F(t,\bar{t}')]}{\sqrt{\Lambda(t,\bar{t}')F(t,\bar{t}')}};$$

$$\Lambda(t,\bar{t}') = \sqrt{\kappa^2 + K'^2};\quad F(t,\bar{t}') = \sqrt{\frac{\ddot{\boldsymbol{K}}'^2}{2}}.$$

The sum in (14) implies summation over all solutions of equation (10) (for simplicity we do not introduce the index numbering these solutions).

Assuming that the pre-exponential function is a slowly varying function, we estimate integral (14) by the stationary phase method. The points of the stationary phase are found from the solution of equation

$$\frac{\boldsymbol{K}^2}{2} - \frac{1}{2}(\boldsymbol{K}'^2 + \kappa^2)\frac{\mathrm{d}\bar{t}'}{\mathrm{d}t} = E,\quad E = \Omega - I_{\mathrm{p}}, \tag{15}$$

where $\boldsymbol{K}(t) \equiv \boldsymbol{K}(t,\bar{t}')$. In order to find the derivative $\mathrm{d}\bar{t}'/\mathrm{d}t$, we differentiate equation (11) with respect to t and obtain

$$\frac{\mathrm{d}\bar{t}'}{\mathrm{d}t} = -\frac{[\ddot{\boldsymbol{K}}'^2]_{t,\bar{t}'}}{\ddot{\boldsymbol{K}}'^2}, \tag{16}$$

where

$$[\ddot{\boldsymbol{K}}'^2]_{t,\bar{t}'} \equiv \frac{\partial^2 \boldsymbol{K}'^2}{\partial t\partial\bar{t}'}.$$

As a result of integration in t, the amplitude A can be expressed as

$$A = \sum_j a_j \exp(\mathrm{i}S_{\mathrm{cl}} + \mathrm{i}\Omega\bar{t}_j), \tag{17}$$

where

$$a_j = \frac{2\pi}{\mathrm{i}^{1/2}}\frac{f(\bar{t}_j,\bar{t}'_j)}{\sqrt{\Lambda_j F_j \delta_j}}\exp(-\Lambda_j^3/3F_j); \tag{18a}$$

$$\Lambda_j = \Lambda(\bar{t}_j,\bar{t}'_j);\quad F_j = F(\bar{t}_j,\bar{t}'_j); \tag{18b}$$

$$\delta_j = \boldsymbol{K}(\bar{t}_j,\bar{t}'_j)\left[\frac{\partial \boldsymbol{K}(\bar{t}_j,\bar{t}'_j)}{\partial\bar{t}_j}\right], \tag{18c}$$

and the pair of real times $\{\bar{t}_j,\bar{t}'_j\}$ is the jth solution of the system of equations

$$\dot{\boldsymbol{K}}'^2 - \frac{1}{3}\frac{(\kappa^2 + \boldsymbol{K}'^2)}{\ddot{\boldsymbol{K}}'^2}\dddot{\boldsymbol{K}}'^2 = 0, \tag{19a}$$

$$\boldsymbol{K}^2 + (\boldsymbol{K}'^2 + \kappa^2)\frac{[\ddot{\boldsymbol{K}}'^2]_{t,\bar{t}'}}{\ddot{\boldsymbol{K}}'^2} = 2E. \tag{19b}$$

Note that the result of (17) coincides in form with (4), but in (4) the summation is performed over quantum (complex) trajectories, and in (17) – along classical (real) trajectories. Thus, expression (17) is a representation of the amplitude of harmonic generation by means of partial amplitudes associated with classical closed trajectories. In the next section, we present a comparison of the numerical results obtained with the help of systems (5) and (19) (hereinafter referred to as 'saddle' and 'tunnel' solutions, respectively). Of practical interest are solutions of the system of equations (19), in which quantum corrections are neglected, i.e., those satisfying equations

$$\dot{\boldsymbol{K}}'^2 = 0, \tag{20a}$$

$$\boldsymbol{K}^2 = 2E \tag{20b}$$

(below we will denote these solutions as 'classical').

3. Numerical results

Numerical calculations for pairs of times t_j, t_j' and their corresponding values of the classical action determining the HHG amplitude were carried out for linearly polarised and bicircular fields. In the case of linear polarisation, the vector potential has the form

$$\boldsymbol{A}(t) = \frac{\mathrm{d}}{\mathrm{d}t}\boldsymbol{R}(t),\quad \boldsymbol{R}(t) = \frac{\boldsymbol{F}}{\omega^2}\exp\left(-2\ln 2\frac{t^2}{T^2}\right)\cos\omega t, \tag{21}$$

where $\boldsymbol{F} = \boldsymbol{e}_x F$ is the field strength; ω is the carrier frequency; $T = 2\pi N/\omega$ is the effective pulse duration; and N is the number of field periods. In the case of a bicircular field, the vector potential is written in the form

$$\boldsymbol{A}(t) = \frac{\mathrm{d}}{\mathrm{d}t}\boldsymbol{R}(t),\quad \boldsymbol{R}(t) = \boldsymbol{R}_1(t) + \boldsymbol{R}_2(t), \tag{22}$$

$$\boldsymbol{R}_i(t) = \frac{F}{\omega_i^2}\exp\left(-2\ln 2\frac{t^2}{T^2}\right)(\boldsymbol{e}_x\cos\omega_i t + \boldsymbol{e}_y\eta_i\sin\omega_i t), \tag{23}$$

where $\omega_1 = \omega$, $\omega_2 = 2\omega$; $T_i = 2\pi N/\omega_i$; $\eta_1 = 1$; $\eta_2 = -1$; and $i = 1, 2$. Numerical calculations were performed for the carrier frequency $\omega = 0.775$ eV, the intensity $I = cF^2/(8\pi) = 10^{14}$ W cm^{-2}, $N = 3$ and $I_{\mathrm{p}} = 13.65$ eV. The characteristic values of the ponderomotive energy for the linearly polarised field are $u_{\mathrm{p}}^{(\mathrm{lin})} = F^2/(4\omega^2) = 23.9$ eV and in the case of the bicircular field $u_{\mathrm{p}}^{(\mathrm{bc})} = F^2/(2\omega_1^2) + F^2/(2\omega_2^2) = 5F^2/(8\omega^2) = 59.76$ eV. Figures 1 and 2 show the calculation results for a linearly polarised field and a bicircular field, respectively.

As can be seen from the calculation results (see Figs 1a and 2a), an electron is released from the atom every half-cycle for a linearly polarised field [5, 8] and every third of the period for a bicircular field [19, 20]. It is obvious that the ionisation process is most effective in a time interval for which the peak intensity is commensurable with the maximum in the laser pulse (in our case this interval is defined as $-T < t < T$). Note that in the case of linear polarisation, ionisation occurs practically at the maximum of the square of the field strength, whereas in the case of a bicircular field, a significant shift is observed from the instants of time in which the square of the field strength is maximal. As our analysis shows, this time shift is due to a nonzero initial energy at the moment of ionisation in the bicircular field.

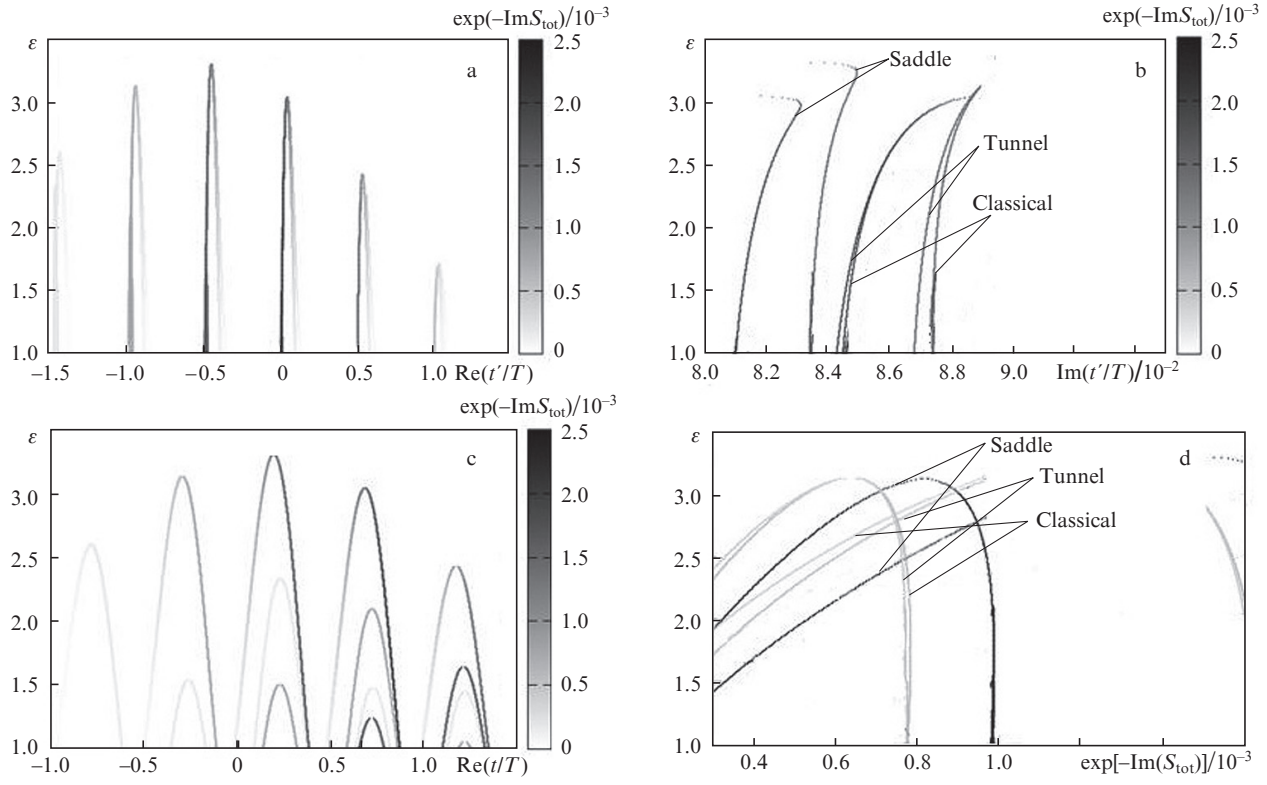

Figure 1. Dependences of (a) $\mathrm{Re}\,(t'/T)$, (b) $\mathrm{Im}\,(t'/T)$, (c) $\mathrm{Re}\,(t/T)$ and (d) $\exp(-\mathrm{Im}S_{\mathrm{tot}})$ on the reduced energy $\varepsilon = E/u_{\mathrm{p}}^{(\mathrm{lin})}$ for a linearly polarised laser field. Curves (a – b) have a gradient coloration in accordance with the values of $\exp(-\mathrm{Im}S_{\mathrm{tot}})$[see $S_{\mathrm{tot}}(t, t')$ in (2)]. The solutions corresponding to systems (5) ('saddle'), (19) ('tunnel') and (20) ('classical') in Figs 1a and 1c are indistinguishable. Figures 1b and 1d show numerical results for two solutions that determine the largest contribution to the amplitude A.

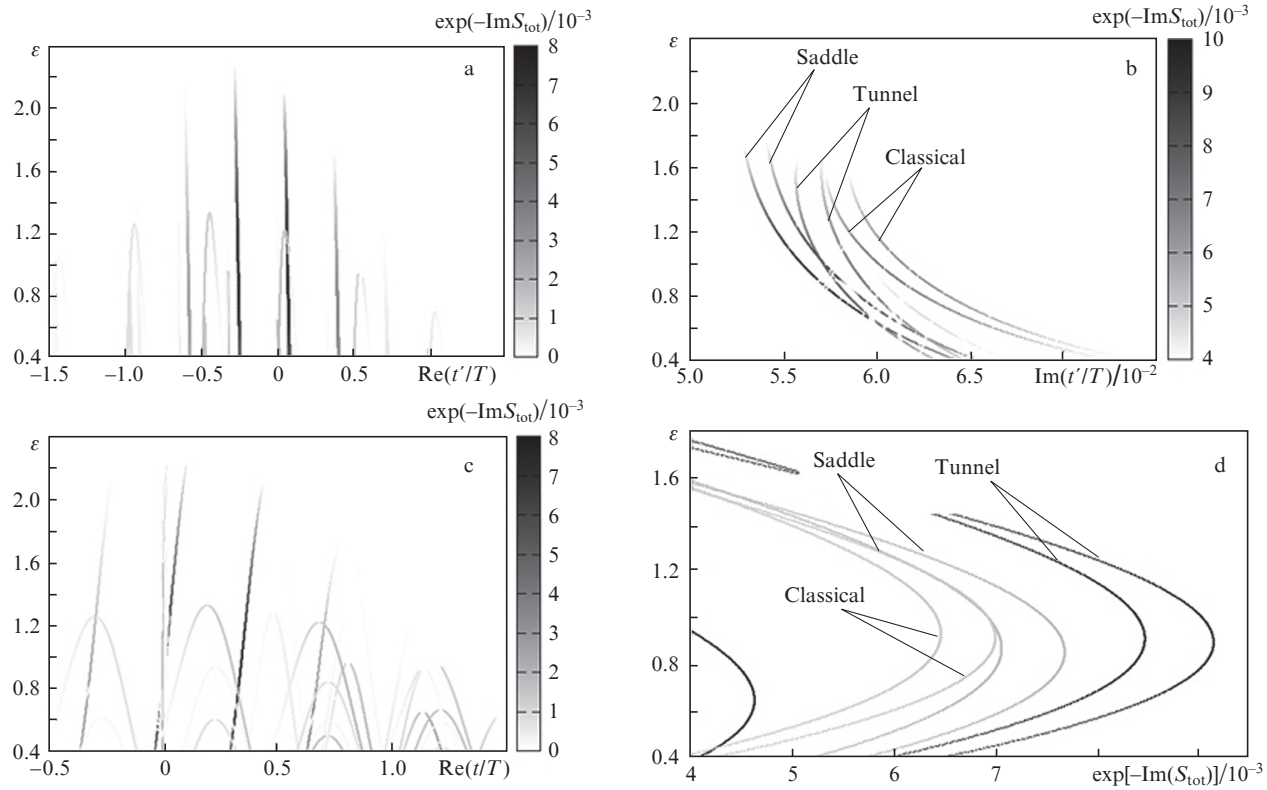

Figure 2. Same as in Fig. 1, but for a bicircular field. The reduced energy is defined as $\varepsilon = E/u_{\mathrm{p}}^{(\mathrm{bc})}$.

In contrast to a linearly polarised field, for which a return to an atomic core with a certain energy is possible with almost the same probability along a long ($\bar{t}_j - \overline{t_j'} > 0.65T$) and short ($\bar{t}_j - \overline{t_j'} < 0.65T$) trajectories [5] (see Fig. 1c), in the case of a bicircular field, the return is most probable only along a short trajectory ($\bar{t}_j - \overline{t_j'} < 0.4T$) [19] (see Fig. 2c). Suppression of motion along a long trajectory is caused by an extremely low probability of the exit of an atomic electron into the continuum. The above analysis shows that the dynamics of the electron (the process of rescattering) in linearly polarised and bicircular fields is significantly different, so that the identification of analogies in electron rescattering in these fields [21] upon transition to a rotating coordinate system [22] is not consistent with physical and mathematical points of view.

According to numerical results, the real parts for the recombination times obtained from the solution of systems (5), (19) and (20) do not differ significantly from each other. Let us now analyse the results for the imaginary parts t', which determine the imaginary part of the action $S_{cl}(t, t')$. Figures 1b and 1d and 2b and 2d show the dependences of the imaginary part t' and $\exp(-\mathrm{Im}S_{tot})$ on the reduced energy E. The imaginary parts t', corresponding to Fig. 1b are 'classical' and 'tunnel' solutions, were calculated with the help of relation (9) and real roots obtained from the solution of the systems of equations (20) and (19). The deviation of the two solutions from the 'saddle' solution does not exceed 10%. The accuracy of the approximate solutions decreases dramatically in the energy region in which the second derivative $S_{cl}(t, t')$ with respect to t is close to zero. Indeed, following the definition of $S_{cl}(t, t')$ [see (2)], we have

$$\frac{\partial^2 S_{cl}}{\partial t^2} = -\frac{\partial \boldsymbol{K}^2}{\partial t}, \tag{24}$$

i.e., in this energy region an extremum (maximum) of the particle energy is realised at the time of return (rescattering) $\boldsymbol{K}^2$ (see, for example, the maxima in Fig. 1c). In the vicinity of these energies, a caustic of 'short' and 'long' trajectories is observed [23], and the solution of system (5) changes drastically: for example, the imaginary part of time t' experiences a sharp bend (see Figs 1b and 2b), whereas the value of the imaginary part t' sharply increases. In this energy region, the solutions lie in the vicinity of the Stokes line [24] and a small change in E leads to a jump-like change in the asymptotic behaviour of the partial amplitude A_j [determined by the pair of solutions $\{t_j', t_j\}$ on the jth branch of the solution of system (5)], which consists in changing the oscillation (or smoother) dependence by a sharp exponentially similar damping. It is obvious that the proposed method for estimating the HHG amplitude and calculating the times $\{t, t'\}$ is not suitable for the caustic region and requires special consideration [25–28]. For small energies E, that is, for $\Omega \sim I_p$, the accuracy of the solutions of system (20) deteriorates significantly. Indeed, in this case the classical mechanism of the electron energy exchange in the continuum ceases to be dominant in comparison with the quantum one; therefore, the quantum corrections in this energy region turn out to be significant.

Figures 1d and 2d show the dependence of $\exp(-\mathrm{Im}S_{tot})$ on the reduced energy $\varepsilon = E/u_p^{(lin)}$ (in the case of linear polarisation) and $\varepsilon = E/u_p^{(bc)}$ (for a bicircular field). Qualitatively, the behaviour of $\exp(-\mathrm{Im}S_{tot})$ for all three types of solutions is the same. Moreover, the dependence of $\exp(-\mathrm{Im}S_{tot})$ on ε for 'tunnel' and 'classical' solutions reduces to a 'saddle' solution by simple scaling: for example, with high accuracy we can assume that $\exp(-\mathrm{Im}S_{tot})$ for the 'saddle' solution corresponds to $1.2\exp(-\mathrm{Im}S_{tot})$ for the 'tunnel' solution. This simple scaling of the two results is due to the contribution of higher corrections in $\Delta t'$ to the imaginary part of the classical action S_{cl}; however, their calculation requires a higher degree of expansion of equation (5a) in a series in $\Delta t'$ and is beyond the scope of this paper.

4. Conclusions

A method is proposed for estimating the HHG amplitude by atoms in an intense laser field with two spatial components. The method based on the analysis of quasi-classical equations at the moments of transition of a bound electron to a laser-modified continuum and its subsequent recombination. Using the smallness of the time of subbarrier motion of the electron with respect to the characteristic period of the laser field, it is shown that it is possible to develop a formalism in which the harmonic generation amplitude can be represented as a sum of partial amplitudes associated with classical closed trajectories. The classical equations and quantum corrections to them are obtained, which determine the classical (real) times of the beginning and the end of motion along closed classical trajectories. By the example of linearly polarised and bicircular fields, the accuracy of calculating the ionisation and recombination times is determined in the framework of the developed formalism. It is shown that the developed formalism makes it possible to abandon the physically unreasonable concept of 'missed' trajectories in the analysis of generation of high harmonics and perform an analysis within the framework of closed classical trajectories, as, for example, in the case of a linearly polarised field.

In conclusion, we note that the presented formalism can potentially be extended to problems of harmonic generation with allowance for the Coulomb potential, for example, using an approach that uses the classical trajectories modified by the Coulomb interaction [18, 29, 30]. To date, the inclusion of the Coulomb interaction of an optical electron in the generation of harmonics has been accomplished using the heuristic approach [27, 31–33], but the accuracy and correctness of such a generalisation require additional studies.

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