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IN MEMORY OF LEONID VENIAMINOVICH KELDYSH

Multiphoton ionization by a very short pulse

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Abstract. The detachment of a bound electron by an electric field pulse whose duration ranges from a fraction of to a few times the optical cycle but is long compared to $\hbar/I$ ($\hbar$ — Planck constant, I — binding energy) is studied theoretically, simulating the ionization of atoms by extremely short laser pulses. Because of the strong nonlinearity, the solution to the problem does not reduce to the sum of monochromatic harmonic contributions and depends significantly on the pulse shape features. A general analysis is carried out for an analytical pulse shape, and exact formulas are given for standard pulse shapes such as solitonlike, gaussian, lorenzian, etc., one or a half optical cycle in duration. The intensity and pulse length dependences of the ionization probability are of a near-universal tunneling type at high intensities. However, at moderate intensities in the multiphoton regime, these dependences differ widely for different pulse shapes, with ionization probabilities always a few orders of magnitude higher than for ionization by a monochromatic wave of the same intensity and mean frequency.

Keywords: multiphoton processes, tunnel and multiphoton ionization, relativistic ionization, very short laser pulses, intense laser radiation

Remarkable progress in generating very high intensity laser fields was closely related to a corresponding reduction in pulse duration [1, 2]. Therefore, actual multiphoton processes, i.e., those requiring the simultanious participation of many ($\gg 1$) photons, are observed typically in experiments with ultrashort pulses (USPs). With this reduction continuing, pulselengths become comparable to the optical field cycle duration [3–8]. Under such conditions, the usual concept of transition (ionization) probability per unit time makes no sense. The only meaningful quantity remains the total — after the whole pulse — transition probability. Moreover, the frequency spectrum of the pulses under consideration is very broad and, because of the extreme nonlinearity of the process, its probability does not reduce to the sum of independent harmonics contributions.

The physical essence of ionization process in the high intensity USP case may be thought of as an interaction of and competition among the contributions of many harmonies, depending not only on the spectrum, but also on the phase relations of different harmonies, i.e., higher order field correlations. In other words, this means that the result is very sensitive to the exact pulse shape. In this article, some extreme particular cases are theoretically studied, corresponding to USPs a few or even half an optical cycle long.

Recently, a few groups have investigated both experimentally and theoretically an even more extreme limiting case, in a sense — the ionization of atoms by a pulse much shorter than characteristic electron times (the inverse optical transition frequency between neighbouring energy levels). They realized these conditions experimentally with alkali atoms, excited to very high Rydberg states, corresponding to quasiclassical electron motion and small interlevel distances. A field pulse acted in such a case as an (quasi)instantaneous kick, moving electron from one — bounded — Kepler orbit to another — unbounded.

In contrast to this, the problem discussed below is essentially a quantum one: ionization from the tightly bound state, e.g., the ground state, by a pulse one or one-half optical cycle long, but much longer than the 'atomic cycle' — $\hbar/I$, $\hbar$ being the Planck constant and I the ionization energy. This means that the average energy of a photon in the pulse is small compared to the ionization energy. For an atomic electron, this is slowly varying perturbation, and therefore the same adiabatic treatment can be applied to this problem which was exploited earlier [9] for ionization by intense monochromatic waves. The basis for this is the observation that the final — free — state of an electron in the process under consideration is much more sensitive to such type of perturbations than the initial one — strongly bound and localized. So, the transition probability is calculated as that of a first order transition from the unperturbed initial atomic state to the final 'exact' state of the free electron in the strong time-dependent electric field. The latter of these states accounts for the field action nonperturbatively and contains the main contribution to the transition amplitude. For the fields below atomic, i.e., intensities up to the PW/cm^2 range, the most (and only) important defect in this approach is neglecting electron–ion Coulomb interaction in the final state, i.e., a Born-type approximation. The significance of such approximate solutions may seem questionable now. This kind of quantum problem — a single electron in an external field, including both atomic and electromagnetic — is certainly within the limits of modern computing abilities. During the last decade, several algorithms have been proposed and successfully applied to the problems of multiphoton ionization and some other related ones, such as UV higher harmonics generation. Still, analytic solutions, even semiquantitatively correct, also have their advantages, not being restricted by any definite set of parameter values. They may be useful in representing an

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overall view of the process and trends due to variation of parameters, or as a starting point in analyzing more complicated, e.g., multielectron, systems.

Let the spatially uniform time-dependent electric field $\mathbf{F}(t)$ be given by

$$\mathbf{F}(t) = \mathbf{F} f'(\omega t)\,. \tag{1}$$

Here, $f'(x)$ is the derivative of the function f over its argument x, and ω is the inverse characteristic timescale of the pulse. In such a field, the wave function of the free electron is

$$\psi_{\mathbf{p}}(\mathbf{r}, t) = \exp\left[\frac{\mathrm{i}}{\hbar}\left(\mathbf{p}(t)\,\mathbf{r} - \int_0^t \frac{p^2(t')}{2m}\,\mathrm{d}t'\right)\right] \tag{2}$$

with

$$\mathbf{p}(t) = \mathbf{p} + \frac{e\mathbf{F}}{\omega} f(\omega t)\,. \tag{3}$$

Following the usual first-order perturbation theory, the transition probability from the initial state $\psi_0(\mathbf{r}) \exp\left[(\mathrm{i}/\hbar)\, It\right]$ to the final state $\psi_{\mathbf{p}}(\mathbf{r}, t)$ can be calculated:

$$w_{i\mathbf{p}} = \frac{e^2\mathbf{F}^2}{\hbar^2\omega^2} \times \left|\int_{-\infty}^{\infty} \mathrm{d}x\, R_{\parallel}\left(\mathbf{p} + \frac{e\mathbf{F}}{\omega} f(x)\right) \exp\left(\mathrm{i}\,\frac{I}{\hbar\omega}\,\Phi(x)\right)\right|^2, \tag{4}$$

with phase function $\Phi(x)$ defined as

$$\Phi(x) = \frac{1}{I}\int_0^x \left(I + \frac{p^2(x')}{2m}\right)\mathrm{d}x' - \mathrm{i}\,\frac{\hbar\omega}{I}\ln f'(x)\,. \tag{5}$$

Here, $R_{\parallel}(\mathbf{p})$ is the transition matrix element of the coordinate component parallel to the field $\mathbf{F}$,

$$R_{\parallel}(\mathbf{p}) = \int \exp\left[-\left(\frac{\mathrm{i}}{\hbar}\right)\mathbf{p}\mathbf{r}\right]\mathbf{n}\mathbf{r}\,\psi_0(\mathbf{r})\,d^3r\,,$$

and $\mathbf{n}$ is the unit vector in the field direction.

In order to make the following analysis more vivid, it is convenient to use the representation of all quantities involved in the natural 'atomic' scale, i.e., to define

$$\Omega = \frac{\hbar\omega}{I}\,,\quad \mathbf{q} = \frac{\mathbf{p}}{\sqrt{2mI}}\,,\quad \mathcal{E} = \frac{e\hbar\mathbf{F}}{\sqrt{2mI^3}}\,. \tag{6}$$

Certainly, this means the corresponding transformation of coordinate and time scales. Then, the dimensionless matrix element should be defined as

$$M(\mathbf{q}) = \left(\frac{\sqrt{2mI}}{\hbar}\right)^{5/2} R_{\parallel}(\mathbf{p})\,. \tag{7}$$

According to the above claim, the whole consideration in this article is for $\Omega \ll 1$.

The crucial parameter of the theory is then the ratio of dimensionless field to the frequency:

$$\lambda = \frac{|\mathcal{E}|}{\Omega}\,, \tag{8}$$

which is exactly inverse to the parameter γ introduced in [9] (if ω is considered a characteristic frequency of the process).

The factor $1/\Omega$ being the large parameter of the theory, the integral in (4) can be calculated by the stationary phase method. The stationary phase point(s) in the complex variable x plane is found from the equation

$$\left.\frac{\partial\Phi(x, \mathbf{q})}{\partial x}\right|_{x_s} = 1 + \left(\mathbf{q} + \mathbf{n}\,\lambda f(x_s)\right)^2 - \mathrm{i}\Omega\,\frac{f''(x_s)}{f'(x_s)} = 0\,. \tag{9}$$

Then, the transition probability

$$w_{i\mathbf{p}} = 2\pi\Omega\lambda^2\left|\sum_s \frac{M\left(\mathbf{q} + \mathbf{n}\lambda f(x_s)\right)}{\sqrt{|\Phi''(x_s, \mathbf{q})|}} \exp\left(-\frac{\mathrm{i}}{\Omega}\,\Phi(x_s, \mathbf{q})\right)\right|^2, \tag{10}$$

with summation over all saddlepoints x_s. Contributions of different saddlepoints are exponentially different and only the dominating one must be kept in (10). Generally, there is one such dominating saddle point — that corresponding to the lowest value of the positive imaginary part of $\Phi(x_s, \mathbf{q})$. However, in many cases, due to some symmetry of the pulse function $f(x)$, pairs or groups of equivalent saddle points exist with equal values of $\mathrm{Im}\,\Phi(x_s, \mathbf{q})$ but different phase factors $\mathrm{Re}\,\Phi(x_s, \mathbf{q})$. Interference of their contribution results in oscillations of the ionization probability as a function of pulse parameters λ and Ω.

Considered as a function of its argument $\mathbf{q}$, this probability is the momentum distribution function of emitted electrons. Note, however, that $\mathbf{q}$ in this formulae is momentum at the time instant when $f(x) = 0$. So, if $f(\infty) \neq 0$, as it is, e.g., in examples 1 and 4 below, the momentum distribution of ejected electrons is distribution (10) but shifted by $\delta\mathbf{q} = \mathbf{n}\lambda f(\infty)$, as is done in formulae (17) and (43) for the above mentioned examples 1 and 4.

Typically, distribution is Gaussian around some average momentum $\mathbf{q}_{\mathrm{m}}$, to be defined from the condition of minimum of $\mathrm{Im}\,\Phi(x_s, \mathbf{q})$, which, accounting for (9), reduces to

$$q_{\parallel\,\mathrm{m}} = -\frac{\lambda}{x''_{\mathrm{sm}}}\,\mathrm{Im}\left[\int_0^{x_{\mathrm{sm}}} \mathrm{d}x f(x)\right] \tag{11}$$

and $\mathbf{q}_{\perp} = 0$, with $q_{\parallel}$ and $\mathbf{q}_{\perp}$ being the momentum components parallel and perpendicular to the field direction, $x_{\mathrm{sm}} \equiv x_s(\mathbf{q}_{\mathrm{m}})$, and being the x''_s — imaginary part of x_s.

In the vicinity of this sharp maximum, taking into account (9) and (11), the imaginary part of $\Phi(x_s, \mathbf{q})$ can be transformed into

$$\mathrm{Im}\,\Phi(x_s, \mathbf{q}) = x''_{\mathrm{sm}} + \mathrm{Im}\left\{\int_0^{x_{\mathrm{sm}}} \left[\lambda^2 f^2(x) - q^2_{\parallel\mathrm{m}}\right]\mathrm{d}x + x_{\mathrm{sm}} q_{\perp}^2 + \left[x_{\mathrm{sm}} - \mathrm{i}(\lambda f'(x_{\mathrm{sm}}))^{-1}\right](q_{\parallel} - q_{\parallel\mathrm{m}})^2\right\}, \tag{12}$$

with halfwidths defined by the second derivatives of the exponent in (10) over components of the momentum.

A comment should be made about the pre-exponential factor in (10). In deriving this formula, the matrix element $M\left(\mathbf{q} + \mathbf{n}\lambda f(x)\right)$ was treated as a regular function, slowly varying in the vicinity of x_s: $M(\mathbf{q}) \simeq M_0 \equiv M(0)$. However, typically $M(\mathbf{q})$ contains a pole — a singularity $M(\mathbf{q}) = M_0/(1 + q^2)$, in the momentum complex plane [9]. In the whole range of nonlinear absorbtion $\lambda \gg \lambda_{\mathrm{c}}$, with λ_{c} defined below by (46), terms in $\Phi(x_s)$ proportional to λ or λ^2 are much larger than the last term $\sim \Omega$. This pole then comes

very close to the position of the saddle point. This modifies slightly the evaluation of the integral in (4): instead of the saddle point contribution, we have half of the residue at that point, which enhances the pre-exponent in (10) by the factor $\pi/(4\mathcal{E}|f'(x_s)|)$. If two (or a few) equivalent saddle points (and poles of M) are present in (10), each contribution to the transition amplitude must be multiplied by $\operatorname{sign}\operatorname{Im}[f(x_s)]\,[\pi/(4\mathcal{E}|f'(x_s)|)]^{1/2}$. However, strictly speaking, these corrections to the preexponential factor (also like the one discussed below and due to violation of the standard stationary phase method in the vicinity of the singularity in the pulseshape function itself) must be ignored: the pre-exponential factor in (10) and some following formulae should be considered correct only by the order of magnitude, because of the abovementioned Born-type approximation.

Results for a few particular but quite representative examples are shown below.

1. Solitonlike half-cycle pulse (HCP)

$$f(x) = \tanh x \tag{13}$$

which means for electric field strength

$$\mathcal{E}(t) = -\frac{\mathcal{E}}{(\cosh \omega t)^2}\,. \tag{14}$$

Momentum-resolved ionization probability

$$w_{\mathrm{i}}(\Omega,\lambda,\mathbf{q}) = \frac{\pi\Omega}{\sqrt{\Omega^2+\lambda^2}}\,(\lambda^2+\zeta^2)\,|M_0|^2 \times \exp\left\{-\frac{2}{\Omega}\left[(1+\lambda^2)\arctan\frac{\zeta}{\lambda} - \lambda\zeta + \arctan\frac{\zeta}{\lambda}\,(\mathbf{q}-\mathbf{n}\lambda)^2\right]\right\} \tag{15}$$

with parameter

$$\zeta = \frac{1}{\lambda}\left[\sqrt{\Omega^2+\lambda^2}-\Omega\right]. \tag{16}$$

Accounting for the above-mentioned pole in the transition matrix element, formula (15) should be modified to

$$w_{\mathrm{i}}(\Omega,\lambda,\mathbf{q}) = |\pi M_0|^2 \times \exp\left[-\frac{1}{\Omega}\left((1+\lambda^2)\,\pi + 2\arctan\frac{\zeta}{\lambda}\,(\mathbf{q}-\mathbf{n}\,\lambda)^2\right)\right] \times \sinh^2\left[\frac{1}{\Omega}\left((1+\lambda^2)\arctan(\lambda)+\lambda\right)\right]. \tag{17}$$

Moreover, formulae (15) and (17) are derived by the stationary phase method applied to evaluate the integral in (4). However, for $f(x) = \tanh x$ with field decreasing, the saddle point approaches $\mathrm{i}\pi/2$ — the singularity of $f(x)$ itself (not that of the matrix element). In the linear absorption regime, $\lambda < \Omega \ll 1$, this violates conditions of the stationary phase method applicability, also modifying the numerically pre-exponential factor. In the framework of the general analysis below, the exact (in the Born approximation) formulae will be derived that are also valid for a weak field limit. Coinciding with (15) and (17) in the nonlinear field range, for weak fields they contain correction factors S^{reg} for (15) and S^{sing} for (17), represented in formulae (52) and (54).

2. Solitonlike one-cycle pulse (OCP)

$$f(x) = -\frac{3\sqrt{3}}{4\cosh^2 x}\,. \tag{18}$$

The numerical factor is introduced to normalize $|f'(x_{\mathrm{m}})|$ to unity at both extrema of the field strength.

Momentum-resolved ionization probability

$$w_i(\Omega,\lambda,\mathbf{q}) = 8|2\pi M_0|^2 \exp\left[-\frac{\pi}{\Omega}\,(1+q^2)\right] \times\left[1-\cos\left(\frac{2}{\Omega}\operatorname{Re}\Phi(x_s,\mathbf{q})\right)\right]\sinh^2\left(-\frac{1}{\Omega}\operatorname{Im}\tilde{\Phi}(x_s,\mathbf{q})\right), \tag{19}$$

for which

$$\operatorname{Im}\tilde{\Phi}(x_s,\mathbf{q}) \equiv \operatorname{Im}\Phi(x_s,\mathbf{q}) - \frac{\pi}{2}\,(1+q^2) = (1+q^2)\left(x_s''-\frac{\pi}{2}\right) - \frac{1}{6}\left[(5q_\parallel - 2\tilde{\lambda})\,\eta + \sqrt{1+q_\perp^2}\,\xi\right], \tag{20}$$

$$\operatorname{Re}\Phi(x_s,\mathbf{q}) = (1+q^2)\,x_s' - \frac{1}{6}\left[(5q_\parallel - 2\tilde{\lambda})\,\xi - \sqrt{1+q_\perp^2}\,\eta\right]. \tag{21}$$

Saddle point x_s defined by

$$x_s'' \equiv \operatorname{Im} x_s = \frac{\pi}{2} - \frac{1}{2}\arccos\frac{\sqrt{1+q_\perp^2+(\tilde{\lambda}-q_\parallel)^2}-\tilde{\lambda}}{\sqrt{1+q^2}}\,, \tag{22}$$

$$x_s' \equiv \operatorname{Re} x_s = \frac{1}{2}\tanh^{-1}\left[\frac{\xi}{\tilde{\lambda}+\sqrt{1+q_\perp^2+(\tilde{\lambda}-q_\parallel)^2}}\right], \tag{23}$$

field parameter $\tilde{\lambda} = (3\sqrt{3}/4)\,\lambda$ and

$$\xi = \sqrt{2\tilde{\lambda}\left[\sqrt{1+q_\perp^2+(\tilde{\lambda}-q_\parallel)^2}+\tilde{\lambda}-q_\parallel\right]}\,, \tag{24}$$

$$\eta = \sqrt{2\tilde{\lambda}\left[\sqrt{1+q_\perp^2+(\tilde{\lambda}-q_\parallel)^2}-\tilde{\lambda}+q_\parallel\right]}\,. \tag{25}$$

Formula (19) is presented in the form corresponding to the singular matrix element $M(\mathbf{q})$ as described above. The function sinh(...) in (19) accounts for contributions of two pairs of poles (saddle points): one pair with $x_s'' < \pi/2$ and another symmetrically above $\pi/2$. The contribution of the latter pair is significant only at the weakest fields $\lambda \ll \Omega^2$. Thanks to this, formula (19) describes correctly (up to numerical factor ~ 1) the linear absorption. In the whole nonlinear range $\lambda \gg \Omega^2$, this contribution is negligible and sinh does not differ from half of the exponential function of the same argument.

Momentum $q_{\parallel\mathrm{m}}$, corresponding to the distribution function maximum, should be found from the equation

$$q_{\parallel\mathrm{m}}\,x_{s\mathrm{m}}''\Big|_{\mathbf{q}_\perp=0} = \eta \tag{26}$$

and substituted into (19)–(25). For small $\lambda \ll 1$, it is approximately $q_{\parallel\mathrm{m}} \approx (2\tilde{\lambda})^{1/2}/\pi$. For large fields $\lambda \gg 1$, its value approaches $2\tilde{\lambda}/3$.

Oscillations in the field and momentum dependences in (19) arise because of interference contributions due to the pair

of saddle points, symmetrical relative to an imaginary axis. In the total — momentum integrated — ionization probability, their amplitude decreases with a field increase as a result of destructive interference of different momenta contributions:

$$W_{\rm i}(\Omega,\lambda)=\sqrt{\frac{2\pi\Omega}{u}}\,\frac{\Omega}{x''_{sm}}\,|M_0|^2\,\exp\left[-\frac{\pi}{\Omega}\,(1+q_{\rm m}^2)\right]\times\left[1-\exp\left(-\frac{2\tilde\lambda}{\pi\Omega}\right)\cos\left(\frac{4\sqrt{2\tilde\lambda}}{3\Omega}\right)\right]\times\sinh^2\left(-\frac{1}{\Omega}\,{\rm Im}\,\tilde\Phi(x_{sm},\mathbf{q}_{\rm m})\right),\tag{27}$$

with

$$u=\left(x''_s+\frac{1}{4}\,\frac{q\xi+\eta}{(1+q^2)\sqrt{1+(\tilde\lambda-q)^2}}\right)_{\mathbf{q}=\mathbf{q}_{\rm m}}.$$

The oscillating term is written here in a form valid only for $\lambda\ll 1$, as for larger fields this term becomes negligible.

3. Gaussian one-cycle pulse (OCP)

$$f(x)=\exp\left(\frac{1-x^2}{2}\right).\tag{28}$$

Corresponding field pulse shape

$$\mathcal{E}(t)=-\mathcal{E}\omega t\,\exp\left(\frac{1-(\omega t)^2}{2}\right).\tag{29}$$

Then,

$$\Phi(x_s,\mathbf{q})=(1+q^2)\,x_s+2\tilde\lambda q_\parallel\,{\rm Erf}\left(\frac{x_s}{\sqrt{2}}\right)+\tilde\lambda^2\,{\rm Erf}\,(x_s)\,.\tag{30}$$

Here, $\tilde\lambda\equiv\sqrt{\rm e}\,\lambda$, ${\rm Erf}\,(x)$ is the error integral

$${\rm Erf}\,(x)=\int_0^x\exp\,(-y^2)\,{\rm d}y\,,$$

$$x_s(\mathbf{q})=\sqrt{\ln\frac{\tilde\lambda^2}{1+q^2}\mp 2{\rm i}\arccos\frac{-q_\parallel}{1+q^2}}.\tag{31}$$

Only saddlepoints in the upper halfplane of x are relevant. Thus, the signs of the roots must be chosen with positive imaginary parts. Therefore, the signs of the real part are different for two saddlepoints. This is just an example of two equivalent saddlepoint interferences.

$$x''_s\equiv{\rm Im}\,x_s=\frac{1}{\sqrt{2}}\times\sqrt{\sqrt{\left(\ln\frac{\tilde\lambda^2}{1+q^2}\right)^2+4\left[\arccos\left(-\frac{q_\parallel}{\sqrt{1+q^2}}\right)\right]^2}-\ln\frac{\tilde\lambda^2}{1+q^2}}\,,\tag{32}$$

$$x'_s\equiv{\rm Re}\,x_s=\mp\frac{1}{\sqrt{2}}\times\sqrt{\sqrt{\left(\ln\frac{\tilde\lambda^2}{1+q^2}\right)^2+4\left[\arccos\left(-\frac{q_\parallel}{\sqrt{1+q^2}}\right)\right]^2}+\ln\frac{\tilde\lambda^2}{1+q^2}}\,.\tag{33}$$

The equation for $\mathbf{q}_{\rm m}$ in this case looks like

$$\mathbf{q}_{\rm m}\int_0^{x''_{sm}}\left[1-\exp\left(-x''_{sm}u+\frac{u^2}{2}\right)\cos\,(x'_{sm}u)\right]{\rm d}u=-\int_0^{x''_{sm}}\exp\left(-x''_{sm}u+\frac{u^2}{2}\right)\sin\,(x'_{sm}u)\,{\rm d}u\,.\tag{34}$$

As x_{sm} itself is a function of $\mathbf{q}_{\rm m}$, this equation, along with (31), a system of two coupled equations defining both x_{sm} and $\mathbf{q}_{\rm m}$.

The imaginary parts of the Erf functions in (30) can also be represented by integrals similar to those in (34). Taking into account (31),

$$\tilde\lambda^2\,{\rm Im\,Erf}\,(x_s)=-\int_0^{x''_s}\exp\,(-2x''_{sm}u+u^2)\times\left[(1-q_{\parallel{\rm m}}^2)\,\cos\,(2x'_{sm}u)+2q_{\parallel{\rm m}}\,\sin\,(2|x'_{sm}|u)\right]{\rm d}u\,.\tag{35}$$

Unlike the general form of (31)–(33), equations (34) and (35) are written for $\mathbf{q}=\mathbf{q}_{\rm m}$.

All these formulae become substantially simplified and more transparent in the 'multiphoton' ($\lambda\ll 1$) and 'tunneling' ($\lambda\gg 1$) parameter ranges. For moderate intensities ($\lambda\ll 1$), momentum-resolved ionization probability

$$w_{i\mathbf{q}}\approx 4\pi\Omega\,\frac{\lambda^2}{\lambda^2+\lambda_{\rm c}^2}\,x''_{sm}\left|M(0)\right|^2\left[1+\cos\frac{\pi-4q_\parallel}{x''_{sm}\Omega}\right]\times\exp\left[-\frac{2}{\Omega}\left((1+q^2)\,x''_{sm}-\frac{1}{2x''_{sm}}\right)\right],\tag{36}$$

with x''_{sm} given by

$$x''_{sm}=\sqrt{\ln\frac{1}{\tilde\lambda^2+\lambda_{\rm c}^2}}\gg 1\,,\tag{37}$$

$$q_{\parallel{\rm m}}\approx-\frac{\pi}{2x''^2_{sm}\,(x''^2_{sm}-1)}\ll 1\,.\tag{38}$$

Strictly speaking, formula (36) is correct for low fields ($\lambda\ll\lambda_{\rm c}\equiv\exp\,[-1/(2\Omega^2)]$, linear absorption) and moderate fields ($\lambda_{\rm c}\ll\lambda\ll 1$). In the intermediate range ($\lambda\sim\lambda_{\rm c}$), it seems to be a reasonable interpolation. Oscillations of transition probability to any particular momentum due to the interference of two saddlepoint contributions are very strong — up to complete cancellation. However in the total (momentum integrated) ionization probability, they are gradually damped with a field increase because of the momentum dependence of their phases,

$$W_{\rm i}=8\,\frac{\lambda^2 x''^2_{sm}}{\lambda^2+\lambda_{\rm c}^2}\left(\frac{\pi\Omega}{2x''_{sm}}\right)^{5/2}|M(0)|^2\times\left[1+\exp\left(-\frac{2}{\Omega x''^3_{sm}}\right)\cos\frac{\pi}{x''_{sm}\Omega}\right]\times\exp\left[-\frac{2}{\Omega}\left(x''_{sm}-\frac{1}{2x''_{sm}}\right)\right].\tag{39}$$

As to the strong field tunneling regime ($\lambda\gg 1$), formulae (48)–(50) are universal for any pulse shape, the only difference being in the particular value of the parameter a — the curvature at the pulse top. For a Gaussian pulse, $a=2$.

4. Lorenzian half-cycle pulse (HCP)

$$f(x) = \arctan x\,. \tag{40}$$

Corresponding field pulse shape

$$\mathcal{E}(t) = \frac{\mathcal{E}}{1+(\omega t)^2}\,. \tag{41}$$

The saddle point is then

$$x_s = \mathrm{i}\tanh\left(\frac{\sqrt{1+q_\perp^2}+\mathrm{i}q_\parallel}{\lambda}\right), \tag{42}$$

and momentum distribution of ionization probability

$$w_\mathrm{i}(\Omega,\lambda,\mathbf{q}) = |\pi M_0|^2 \exp\left[-\frac{2}{\Omega}\left(|x_{sm}| - \lambda^2\,\varphi(|x_{sm}|)\right)\right] \times \exp\left\{-\frac{2}{\Omega}\left[q_\perp^2\,|x_{sm}| + \left(q_\parallel - \frac{\pi\lambda}{2}\right)^2 \times \left(|x_{sm}| + \frac{2}{\lambda}\left(1-|x_{sm}|^2\right)\right)\right]\right\}, \tag{43}$$

with

$$|x_{sm}| = \tanh\frac{1}{\lambda}\,, \tag{44}$$

$$\varphi(x) = \frac{1}{4}\int_0^x \ln^2\frac{1+y}{1-y}\mathrm{d}y\,. \tag{45}$$

The numerical solution of (9) and (11) is strightforward for any reasonable pulseshape. However, a general qualitative analysis is also possible and may be illuminating. There are three essentially different areas in the plane of parameters (Ω,λ):

1. Weak fields and linear absorbtion for $\lambda \ll \lambda_\mathrm{c}(\Omega)$ with λ_c being the effective nonlinearity threshold, substantially dependent on the pulseshape and specified below for some typical pulse shapes. The general definition is

$$\lambda_\mathrm{c}\left|f(x_{s0})\right| = 1\,, \tag{46}$$

with x_{s0} being the root of equation (9) corresponding to $\lambda = 0$. Terms proportional to λ and λ^2 on the right-hand side of (9) can be disregarded. The exponential factor in (10) reduces to an exponentially small amplitude of high frequency harmonics, corresponding to the above-threshold quantum energy $\hbar\omega > I$, always present in the Fourier spectrum of a broadband signal.

2. Nonlinear regime: $\lambda > \lambda_\mathrm{c}$. The last term in (9) can be omitted. Then,

$$x_s = f^{-1}\left(\frac{-q_\parallel \pm \mathrm{i}\sqrt{1+q_\perp^2}}{\lambda}\right). \tag{47}$$

Here, $f^{-1}(y)$ is a function, the inverse of $f(x)$. The sign of the imaginary part of its argument must be fixed so as to correspond to $x'' > 0$.

2a. High fields — $\Omega^{-1} \gg \lambda \gg 1$.

Without any loss of generality, one can always choose the point $x = 0$ to be the absolute maximum of $f'(x)$, i.e., field strength, and $f'(0) = 1$. This last condition just fixes the exact value of λ. If there are a few equivalent maxima, each of them can be treated separately. In the range of interest around this point, $f(x)$ can be approximated by a cubic parabola:

$$f(x) \approx f_0 + x - \frac{1}{6}\,ax^3\,, \quad 0 < a \sim 1\,. \tag{48}$$

Then, after simple calculations,

$$\mathrm{Im}\,\Phi(x_s,\mathbf{q}) = \frac{2}{3\lambda}\left[1 + 4a\,\frac{(\mathbf{q}-\mathbf{q}_\mathrm{m})^2}{\lambda^2}\right], \tag{49}$$

$$q_{\parallel\mathrm{m}} \approx \lambda\left[f(\infty) - f_0\right]. \tag{50}$$

This corresponds in [9] to quasistatic tunneling during a short $\delta x \sim \sqrt{|\mathcal{E}|}$ time interval around the field maximum. The momentum distribution of photoelectrons is Gaussian with halfwidth $\Delta q_\parallel = (\lambda/4)\sqrt{3|\mathcal{E}|/a}$.

2b. $1 \gg \lambda \gg \lambda_\mathrm{c}(\Omega)$ — moderate fields. Compared to the weak and strong field cases, in this one the λ-dependence of $\hat{\mathrm{Q}}(\mathrm{xs},\mathrm{q})$ is more diverse, depending on the details of the pulse shape, particularly singularities of the function $f(x)$ in the upper half-plane of the complex variable x. The Gaussian shape $f(x) \sim \exp(-x^2/2)$ is the particular case with the only singularity of $f(x)$ being the essential one at infinity. However the most typically pulselike function $f(x)$ has singularities (poles, branching points) in the complex plane of variable x at some x_pol with the imaginary part $x''_\mathrm{pol} \sim 1$. Then, for weak fields, just the $\exp(-2x''_\mathrm{pol}/\Omega)$ defines the amplitude of the high frequency Fourier component responsible for single quantum ionization. With the field increasing, the saddle point x_s moves from x_pol to the real axis. Let the singularity closest to the real axis be the k-th order pole, i.e.,

$$f(x) \approx \frac{A}{(x-x_\mathrm{pol})^k}$$

for $|x - x_\mathrm{pol}| \ll 1$. Then, as will be shown below, the ionization amplitude in the whole domain $\lambda \ll 1$, including both weak and moderate field ranges, beside the weak field factor $\exp(-2x''_\mathrm{pol}/\Omega)$, is dependent only on a single parameter,

$$z = \frac{(\lambda A)^{1/k}}{\Omega}\,, \tag{51}$$

and the moderate field range starts at $|z| \sim 1$, i.e., $\lambda_\mathrm{c} \sim \Omega^k$. Note that the first two of the above-described examples are dominated by such singularities, the first one corresponding to $k = 1$ and the second to $k = 2$. The saddle points (and possible poles of matrix element $M\big(\mathbf{q} + \mathbf{n}\lambda f(x)\big)$ coincident with them in the pre-exponential factor)

$$x_s(\mathbf{q}) = x_\mathrm{pol} + \left[\frac{A\lambda}{1+q^2}\left(\pm\mathrm{i}\sqrt{1+q_\perp^2} - q_\parallel\right)\right]^{1/k}, \tag{52}$$

with both signs in the argument being relevant, as all of these $2k$ points are in the close vicinity of x_pol, which itself is in the upper halfplane. However, in the moderate field strength range $|z| > 1$, only one of them dominates — that with the minimal value of x''_s; or, one pair of such points, if, depending on pole order k and $\chi \equiv \arg A$, there are in the whole set (52) such a pair of mirror symmetric relative to the imaginary axis

elements, with the minimal value of the imaginary part. The second example above with $k = 2$ and $\chi = 0$ corresponds to just such a case $(x_s - x_{\rm pol})_{q=0} = \sqrt{\lambda/2}\,(\pm 1 - {\rm i})$. In general,

$$\begin{aligned}{\rm Im}\,\Phi(x_s, \mathbf{q}) &= x''_{\rm pol} - \frac{2k}{2k-1}\,\gamma\,(\lambda|A|)^{1/k} \\ &+ \frac{q_\perp^2}{(\Delta q_\perp)^2} + \frac{(q_\parallel - q_{\parallel {\rm m}})^2}{(\Delta q_\parallel)^2}\,, \end{aligned} \tag{53}$$

with

$$\begin{aligned}&\Delta q_\parallel^2 \approx \Delta q_\perp^2 \approx x''_{\rm pol} + o(\lambda^{1/k})\,, \\ &q_{\parallel {\rm m}} = \frac{\sqrt{1-\gamma^2}}{(2k-1)\,x''_{\rm pol}}\,(\lambda|A|)^{1/k}\,, \\ &\gamma = \max_s \frac{(x_{\rm pol} - x_s)''}{|x_s - x_{\rm pol}|} \end{aligned} \tag{54}$$

with index $s = 1, 2, ..., 2k$ marking different elements of set (52). Thus, in the moderate field strength range, ionization probability increases as $\exp\,[4k\gamma|z|/(2k-1)]$ and the average momentum as $\lambda^{1/k}$. If there is only one dominating saddle point,

$$\begin{aligned}W_i(\Omega, \lambda) &= \sqrt{\pi}\left(\frac{\Omega}{2x''_{\rm pol}}\right)^{3/2} |M_0|^2 \times \\ &\times \exp\left[-\frac{2}{\Omega}\,x''_{\rm pol} + \frac{4k}{2k-1}\,\gamma|z|\right], \end{aligned} \tag{55}$$

and

$$\begin{aligned}W_{\rm i}(\Omega, \lambda) &= \sqrt{\frac{\pi}{2}}\left(\frac{\Omega}{x''_{\rm pol}}\right)^{3/2} |M_0|^2 \\ &\times \exp\left(-\frac{2}{\Omega}\,x''_{\rm pol} + \frac{4k}{2k-1}\,\gamma|z|\right) \\ &\times \left\{1 - \exp\left[-\frac{2}{x''_{\rm pol}\Omega}\left(\frac{\gamma|z|}{2k-1}\right)^2\right]\right. \\ &\times \left.\cos\left(\frac{4k}{2k-1}\,\sqrt{1-\gamma^2}\,|z|\right)\right\}, \end{aligned} \tag{56}$$

if a pair of symmetric saddle points contribute. It should be noted that, in all arguments of exponential and trigonometric functions, only leading terms in $\lambda \ll 1$ are shown in these formulae.

Derived by a stationary phase asymptotic evaluation of integral in (4), formulae (55) and (56) are valid for the moderate field range $\Omega^k \ll \lambda \ll 1$. Their inapplicability for weak fields is clearly seen from the fact that they do not follow the usual $\sim \mathcal{E}^2$ dependence as $\lambda \to 0$. The reason for that was already mentioned above in discussing the second example: at $z < 1$, contributions of all $2k$ saddle points, surrounding $x_{\rm pol}$, become of the same order. It is easy to account for all of them, which would restore the correct $\sim \mathcal{E}^2$ behavior in weak fields. Still it is not the whole story. The numerical coefficient appears to be wrong. The reason is that, besides all these poles and saddle points, the point $x_{\rm pol}$ itself is the essential singularity of the integrand in (4): $\exp\,[{\rm i}(\lambda A)^2 (x - x_{\rm pol})^{-2k+1}/(2k-1)]$. An evaluation of the integral in (4) accounting for this whole structure in the complex plane, valid in the whole domain $|z| \ll 1$, i.e., weak and moderate field ranges, is possible in terms of a fast converging power series in z. The result again is slightly different depending on the presence or absence of the pole in the matrix element. If the matrix element is regular (no pole) and slowly varying, $M(\mathbf{q}) \approx M_0$,

$$\begin{aligned}w_{\rm i}(\Omega, \lambda, \mathbf{q}) &= \lambda^2 |M_0|^2 \exp\left[-\frac{2x''_{\rm pol}}{\Omega}(1+q^2)\right] \\ &\times \left|S_k^{\rm reg}\left(\frac{z^k}{\sqrt{2k-1}}\right)\right|^2, \end{aligned} \tag{57}$$

with

$$S_k^{\rm reg}(y) = 2\pi\sqrt{2k-1}\;y \sum_{n=0}^{\infty} \frac{(-1)^{(k+1)n} y^{2n}}{n!\,[(2k-1)+k]!}\,, \tag{58}$$

and for the case of a singular matrix element,

$$w_{\rm i}(\Omega, \lambda, \mathbf{q}) = \lambda^2 |M_0|^2 \exp\left[-\frac{2x''_{\rm pol}}{\Omega}(1+q^2)\right] \left|S_k^{\rm sing}(z^k)\right|^2, \tag{59}$$

with

$$S_k^{\rm sing}(y) = 2\pi k\,y \sum_{n=0}^{\infty} (-1)^{(k+1)n}\,a_n\,y^{2n}\,, \tag{60}$$

and coefficients a_n defined as

$$a_n = \sum_{m=0}^{n} \frac{(2k-1)^{-m}}{m!\,[(2n+1)\,k - m)]!}\,. \tag{61}$$

The asymptotic form of functions $S_k^{\rm reg}$ and $S_k^{\rm sing}$ at $|z| \gg 1$ exactly coincide with the results of stationary phase calculations in the moderate field regime,

$$S_k^{\rm reg}\left(\frac{z^k}{\sqrt{2k-1}}\right) \approx \sqrt{\frac{\pi k}{|z|}}\,\exp\left(\frac{2k}{2k-1}\,\gamma|z|\right), \tag{62}$$

$$S_k^{\rm sing}(z^k) \approx \pi \exp\left(\frac{2k}{2k-1}\,\gamma|z|\right), \tag{63}$$

and their first terms substituted into formula (51) and (53) give the exact result for the weak field regime. Thus, for a pulse shape with a pole type of singularity, formulae (9)–(12) and (57)–(61) together describe completely the ionization probability for any field strength, restricted only from above by the atomic field, i.e., $\mathcal{E} \ll 1$. However, the weak field regime seems to be of more academic interest: for such short pulses, the effect is hardly experimentally observable.

The last of the above examples corresponds to another type of pulse shape function singularity — the logarithmic branching point ('zeroth order pole').

For long pulses and approximately monochromatic fields, the frequency dependence of true multiphoton process probability is very steep. As the whole consideration above shows, for very short pulses — HCP, OCP, and probably a few ($< 1/\Omega$) cycles-long pulses — it is much slower, though still pretty steep. Qualitatively, this slowing down can be explained as an increase, with the field increasing, of an average effective number n of photons absorbed per single ionization event. Because of a broad frequency spectrum of the pulse, the process is a single-photon one in a weak field and its multiplicity increases gradually to $n \sim \lambda^3$ [9] in the

tunneling regime $\lambda \gg 1$, while in a monochromatic field it is restricted from below, $n > I/(\hbar\omega)$.

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