УДК 537.563.5

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MULTI-PHOTON IONIZATION BY A TWO-COLOR LASER PULSE *

A method of imaginary time [1] is used to calculate the probability of multi-photon ionization of atoms by a femtosecond laser field with an admixture of the second harmonic. The conditions for the second harmonic to dominate over the first harmonic in the course of ionization of atoms are found. It is shown that the average momentum of the photoelectrons ejected from an atom depends on the phase shift between the first and second harmonics, as well as on their mutual polarization. Asymptotic formulas for the ionization rate and the average momentum of photoelectrons are obtained. They generally confirm predictions of a phenomenological model based on the classical views [2].

Keywords: multi-photon ionization, femtosecond laser, terahertz radiation.

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МНОГОФОТОННАЯ ИОНИЗАЦИЯ ИМПУЛЬСОМ ДВУЦВЕТНОГО ЛАЗЕРА

Для вычисления вероятности многофотонной ионизации атомов полем фемтосекундного лазера с добавлением второй гармоники использован метод мнимого времени [1]. Найдены условия преобладания второй гармоники над первой в процессе ионизации атомов. Показано, что средний импульс фотоэлектронов, испущенных атомами, зависит от сдвига фазы между первой и второй гармониками, а так же от их взаимной поляризации. Получены асимптотические формулы для скорости ионизации и среднего импульса фотоэлектронов. Они в целом подтверждают прогнозы феноменологической модели на основе классических подходов [2].

Ключевые слова: многофотонная ионизация, фемтосекундный лазер, терагерцевое излучение.

Multi-photon ionization is though off as a dominant mechanism of the gas ionization and optical breakdown in a focus spot of a femtosecond laser pulse in contrast to the avalanche ionization dominating for longer pulses. Plasma, produced near the focal spot, emits electromagnetic radiation in the frequency range from 1 to 20 THz [3], and the power of the terahertz radiation enhances up to hundreds of times if the laser pulse is partially converted to the second harmonic [4].

Two mechanisms of such an enhancement have been suggested so far.

The first mechanism is based on an intuitively obvious assumption that the multi-photon ionization at the double frequency, 2ω, requires for an atom to absorb two times smaller number of photons to become ionized as compared to the basic frequency, ω. Since the rate of the ionization (in a very rough approximation) is proportional to the intensity of laser radiation (over a large critical value) raised to the num-

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ber of the absorbed photons, the ionization by the 2nd harmonic of the laser field can exceed that by the 1st harmonic, even if the former is much less intensive than the latter.

Another mechanism of the terahertz emission enhancement is based on the observation that the photoelectrons, torn from an atom by the laser field, gain an initial momentum, giving rise to a transient photocurrent [5]. A simple classical model of the phenomenon predicts that the transient photocurrent arises, only if the 2nd harmonic is mixed with the 1st one and only if the phase shift between the harmonics is not multiple to π [2]. However, a strict theory of the terahertz radiation from the focus spot has to be based on a quantum model of the multiphoton ionization.

In this report, an Imaginary Time Method (ITM) is used to evaluate the rate of the multiphoton ionization and generation of the transient photocurrent by a two-color laser pulse composed from the ω and 2ω harmonics. Among the most important parameters of the pulse is the ratio

$$\varepsilon = \frac{E_1}{E_2} \qquad (1)$$

of the 2nd harmonic amplitude E_2 to the amplitude of the 1st harmonic E_1, the phase shift between the harmonics ψ, and their mutual polarization. For the sake of brevity, we consider only the case when both harmonics are linearly polarized along the axis x, so that the electric field of the laser pulse near an atom can be written as

$$E_y = 0,$$
$$E_x = E_1 \cos(\omega t) + E_2 \cos(2\omega t + \psi). \qquad (2)$$

Due to essential nonlinearity of the multiphoton ionization, the contributions of the 1st and 2nd harmonics are not additive. Ionization by a monochromatic field is well described by the theory of L.V. Keldysh [6], developed in the mid of the 1960s. Since it is extremely cumbersome, an imaginary time method (ITM), elaborated mainly by A.M. Perelomov and V.S. Popov [1], was later applied to multi-photon ionization by ultrashort laser pulses [7]. However, a theory suitable for a two-color laser pulse (containing both the 1st and 2nd harmonics) was not formulated. To fill this gap, we get rid of a restriction of ITM, implicitly or explicitly used in earlier works. With respect to the field of the form (2), the restriction meant that ψ must be a multiple of π.

In the framework of ITM, the probability of ionization from the energy level with the binding energy $-I$ by an electromagnetic field within exponential accuracy is determined by the imaginary part of abbreviated action W, evaluated along a classical trajectory, characterized by a given generalized momentum of a bounded electron, finally pulled out from an atom:

$$w_i \propto \exp\left[-\frac{2}{\hbar}\mathrm{Im}(W)\right]. \qquad (3)$$

Assuming the electron to be bounded by short-range forces, the abbreviated action and the trajectory are determined by the laser field, and the abbreviated action has the form [8]

$$W = \int_{t_0}^{t}\left(\frac{m}{2}\mathbf{v}^2(t) + e\mathbf{E}(t)\cdot\mathbf{x}(t) - I\right)dt, \qquad (4)$$

and the trajectory is governed by the classical equations,

$$\frac{d\mathbf{v}}{dt} = \frac{e}{m}\mathbf{E}, \qquad (5)$$

that should be supplemented with the initial conditions,

$$\frac{m}{2}\mathbf{v}^2(t_0) + I = 0, \quad \mathbf{x}(t_0) = 0, \qquad (6)$$

at a complex time $t = t_0$. The initial "time" t_0 should be taken so as to make real the classical trajectory, determined from Eq. (5) and (6), for any real time t. To avoid possible misunderstanding, it is worth noting that some terms that do not change Im[W] have been dropped from Eq. (4).

To simplify notations, we introduce the dimensionless time $\tau = \omega t$ and vector ξ, such that $\mathbf{x} = \left(\frac{eE_1}{m\omega^2}\right)\xi$. Eq. (5) then takes the form

$$\bar{\xi} = \mathbf{F}, \qquad (7)$$

where the dots over ξ denote differentiation with respect to τ, and $\mathbf{F} = \dfrac{\mathbf{E}}{E_1}$ is the dimensionless force. The initial conditions in the new variables read

$$\xi^2(\tau_0) = -\gamma^2, \quad \xi(\tau_0) = 0, \qquad (8)$$

with

$$\gamma = \frac{\sqrt{2mI\omega}}{eE_1}$$

being the Keldysh parameter. In the same notations,

$$w_i \propto \exp\left[-\left(\frac{2I}{\hbar\omega}\right)f_0\right],$$

where

$$f_0 =$$
$$= \mathrm{Im}\left[\frac{2}{\gamma^2}\int_{\tau_0}^{\tau}\left(\xi^2(\tau) + \mathbf{F}(\tau)\cdot\xi(\tau) - \frac{\gamma^2}{2}\right)d\tau\right].$$
(9)

In the limit of multi-photon ionization, $\gamma \to \infty$, (unlike the tunneling ionization at $\gamma \ll 1$), the last term in the right-hand side of Eq. (9) dominates so that $f_0 \approx \mathrm{Im}[\tau_0]$, if all other terms are dropped.

For the linearly polarized field Eq. (2), the trajectory of an electron is one-dimensional since the force $\mathbf{F}$ has only one component,

$$F = \cos(\tau) + \varepsilon\cos(2\tau + \psi).$$
(10)

Ignoring for a while the first of the initial conditions (8), a solution of Eq. (9) with the force (10) can be written in the form

$$\xi = \alpha(\tau - \tau_0) - \left[\cos(\tau) - \cos(\tau_0)\right] -$$
$$- \frac{\varepsilon}{4}\left[\cos(2\tau + \psi) - \cos(2\tau_0 + \psi)\right].$$
(11)

It turns to zero at $\tau = \tau_0$ for any constant α. Value of α should be chosen so as to make both ξ and $\dot{\xi}$ purely real for any real τ. Since

$$\dot{\xi} = \alpha + \sin\tau + \frac{\varepsilon}{2}\sin(2\tau + \psi),$$
(12)

it is evident that α must be purely real. Equating imaginary part of Eq. (11) to zero for an arbitrary real value of τ yields

$$\alpha = \frac{\mathrm{Im}\left[\cos(\tau_0) + \frac{\varepsilon}{4}\cos(2\tau_0 + \psi)\right]}{\mathrm{Im}[\tau_0]}.$$
(13)

Substituting Eq. (12) to the previously omitted first initial condition (8) gives the equation

$$\left[\alpha + \sin(\tau_0) + \frac{\varepsilon}{2}\sin(2\tau_0 + \psi)\right]^2 = -\gamma^2,$$
(14)

that determines the initial "time" τ_0 for a given value of γ. In its turn, α determines average momentum $p = \left(\frac{eE_1}{\omega}\right)\alpha$ of the photoelectrons generated by the laser pulse.

For a particular case of a monochromatic field, $\varepsilon = 0$, Eq. (14) has a purely imaginary root $\tau_0 = i\sinh^{-1}\gamma$ so that $\alpha = 0$, and the ionization probability was calculated by L.V. Keldysh:

$$f_0 = \left(1 + \frac{1}{2\gamma^2}\right)\sinh^{-1}\gamma - \frac{\sqrt{1+\gamma^2}}{2\gamma}.$$
(15)

Another root, $\tau_0 = -i\sinh^{-1}\gamma$, with a negative imaginary part, should be discarded, because it gives an unphysically large rate of ionization.

In a general case, $\varepsilon \neq 0$, Eq. (14) has a lot of roots. Just as with $\varepsilon = 0$, one should select only those of them that result in positive values of f_0. Among these selected roots, one should keep those with smallest positive value of f_0.

Not bothering to analyze all the possibilities, we limit ourselves to the case $\gamma \gg 1$, typical for the experiments with femtosecond lasers. Depending on which of the parameters ε or $\frac{1}{\gamma}$ is larger, there are two limiting cases.

If $\varepsilon \ll \frac{1}{\gamma} \ll 1$, Eq. (14) has an infinite set of roots,

$$\tau_0 \approx i\ln 2\gamma + \pi k - (-1)^k \times$$
$$\times \gamma\varepsilon\left[ie^{-i\psi} + \frac{\sin\psi}{2(\ln 2\gamma - 1)}\right],$$
(16)

corresponding to integer values of k. Since the force of the form (10) is a periodic function of τ, it is sufficient to take only the roots $k = 0$ and $k = 1$. Substituting τ_0 in Eq. (14) gives the value of the parameter

$$\alpha \approx \frac{\varepsilon\gamma^2}{2(\ln 2\gamma - 1)}\sin\psi,$$
(17)

that does not depend on k in the leading order in the parameter ε. The function

$$f_0 \approx \ln 2\gamma - \frac{1}{2} - \frac{2}{3}(-1)^k\varepsilon\gamma\cos\psi,$$

on the contrary, is sensitive to k. Choosing a value of k, leading to the greatest probability of ionization, yields

$$f_0 \approx \ln 2\gamma - \frac{1}{2} - \frac{2}{3}\varepsilon\gamma|\cos\psi|.$$
(18)

Since

$$\frac{I}{\hbar\omega} \gg 1,$$

an admixture of the 2nd harmonic in the laser field can significantly amplify the multi-photon ionization, even if the harmonic is very weak, $\varepsilon\gamma \ll 1$. The amplification effect is described by the last term in (18) and can actually be distinguished experimentally, if

$$\varepsilon\gamma \geq \frac{\hbar\omega}{I}.$$

The amplification of the ionization probability vanishes at $\psi = \pm\frac{\pi}{2}$. On the contrary, the absolute value $|\alpha|$ is maximum at $\psi = \pm\frac{\pi}{2}$. This means that the enhancement of the terahertz radiation from the laser pulse focus due to the 2nd harmonic has two competing mechanisms: amplification of the ionization and the generation of the transient photocurrent. It is possible to determine which of them dominates in a particular experiment by measuring the phase shift ψ, corresponding to the maximum of the radiated power.

In the opposite limit, $\varepsilon \gg \frac{1}{\gamma}$, Eq. (14) yields

$$\tau_0 \approx \frac{i}{2}\ln\left(\frac{4\gamma}{\varepsilon}\right) + \frac{\pi k - \psi}{2} -$$

$$- \frac{1}{2\sqrt{\gamma\varepsilon}}\left[\frac{\sin\left[\dfrac{\pi k + \psi}{2}\right]}{\ln\left(\dfrac{4\gamma}{\varepsilon}\right) - 1} + ie^{\frac{i(\pi k + \psi)}{2}}\right].$$

$$(19)$$

Parameter α is equal to

$$\alpha \approx \frac{\dfrac{\gamma}{\sqrt{\gamma\varepsilon}}}{\ln\left(\dfrac{4\gamma}{\varepsilon}\right) - 1}(-1)^k \sin\left(\frac{\pi k + \psi}{2}\right), \quad (20)$$

and the ionization probability is calculated with the formula

$$f_0 \approx \frac{1}{2}\ln\left(\frac{4\gamma}{\varepsilon}\right) - \frac{1}{4} - \frac{2}{3\sqrt{\gamma\varepsilon}}\cos\left(\frac{\pi k + \psi}{2}\right). \quad (21)$$

For a given ψ, one has to choose a value of k, which falls in the range of

$$-\frac{\pi}{2} < \psi + \pi k < \frac{\pi}{2}.$$

It gives the lowest positive value of f_0, and therefore the greatest probability of ionization.

As in the limit $\varepsilon \ll \frac{1}{\gamma}$, the maximum of the ionization rate is assumed when the phase shift ψ is a multiple of π, whereas the transient photocurrent (proportional to α) is maximum (in absolute value) at $\psi = \pm\frac{\pi}{2}$.

In conclusion, we repeat that adding the 2nd harmonic to the laser pulse is responsible for experimentally observed enhancement of the terahertz radiation from optical breakdown in the focus of a femtosecond laser pulse. The dominant mechanism of the enhancement is the increase of the multi-photon ionization rate. However, the generation of the transient photocurrent also plays a crucial role. In general, the ionization rate is maximum, if the phase shift between the 1st and 2nd harmonics is multiple of π, while the transient photocurrent is maximum at $\psi = \pm\frac{\pi}{2}$. Competition between these two mechanisms can result in maximization of the terahertz radiation emission at intermediate values of the phase shift.

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10.09.2010