Photodetachment of F⁻ by a few-cycle circularly polarized laser field

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Abstract: We report on calculations of the above threshold detachment of F⁻ by a few-cycle circularly polarized laser field, discussing the effects of both the carrier-envelope relative phase and the number of the cycle contained in a pulse on the angular distribution of ejected photoelectron. The results are analyzed in terms of a two-step semiclassical model: after the electrons are detached through tunnelling their motion is determined by the electric field pulse according to the classical dynamics laws. Anisotropies in the angular distributions of the electrons ejected on the plane perpendicular to the laser propagation direction are found that depend on the number of cycle of the laser pulse.

References and Links

1. Sometimes the acronym ATD is used to indicate Above Threshold Dissociation; in the present paper, however, ATD is meant for indicating Above Threshold Detachment.
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1. Introduction

Laser induced above-threshold detachment (ATD) of negative ions has received great interest during past decades[1]. The ATD differs mainly from the above-threshold ionization (ATI) by the absence of the Coulomb attraction of the detached electron by the residual atom. Since the short range nature of the interaction between the atomic core and the outer electron, in experiments carried out for many years with negative ions use has been made of moderately strong laser field enough to observe the nonperturbative effects found in ATI [2]. In early experiments only total detachment rate were recorded[3].

Recently, an image technique [4] has been used to measure energy and angle resolved spectrum of electrons produced by the photodetachment of F^- exposed to a linearly polarized infrared femtosecond laser pulse [5]. In this experiment the spectra exhibit modulations, whose origin has been explained by the Keldysh theory [6]. Through a saddle point analysis of the transition amplitude [7,8], the modulations have been interpreted as the result of interferences of quantum paths leading to the same final state of the detached electron. However, in order to obtain quantitative agreement between the theory and experimental records, numerical simulations have to be performed at peak laser intensities that, generally, result to be higher than the ones estimated [5]. This fact has also been pointed out in ref.s [9, 10], where experimental observations are compared with simulations.

Recently, the photoelectron spectrum of F^- has been measured by exposing the ion to a circularly polarized infrared femtosecond laser pulse [11] containing a large number (almost *20*) of optical cycles. According to the authors of Ref. [11], the main differences observed in the recorded spectrum, as compared to the case of linearly polarized field, is the absence of any structure that can be associated to quantum interference effects [12].

In the present paper we study the ATD of F^- in the presence of a circularly polarized few-cycle laser pulse in the framework of a Keldish-type theory extended to the case of a short laser pulse [13-14]. In particular, we will focus our analysis on the modification of the angular and momenta distributions of the ejected electrons caused by varying the number of optical cycles contained in a single pulse and the envelope-carrier relative phase. The main reason for this analysis stems from the fact that short, high-power laser pulses with duration of few optical cycles are routinely generated and have become available as research tools [15-20]. In fact, it has recently been reported generation of intense, few-cycle laser pulse with a stable carrier envelope phase δ that permits analysis of microscopic motion with extreme precision [16].

As for this kind of pulses the time variation of the laser field depends on δ, triggering and controlling of microscopic processes, as well as their better understanding , may be achieved by varying δ.

2. Basic formulas of F⁻ detachment by a few-cycle pulses.

Let us assume a finite circularly polarized laser pulse with a sin-square envelope, having the following electric field:

$$E(t,\delta)=\begin{cases} \dfrac{E_0}{\sqrt{2}}\,sin^2\left(\dfrac{\pi t}{\tau}\right)\left[\hat{x}\,cos(\omega t+\delta)+\hat{y}\,sin(\omega t+\delta)\right] & t\in[0,\tau] \\ \\ 0 & otherwise \end{cases} \qquad (1)$$

Here τ is the total duration of the pulse, E_0 and ω the field strength and frequency, respectively, and δ the carrier-envelope relative phase. In order to have an integer number of cycles we assume $\tau = n_c T$ with $T=2\pi/\omega$ the period of the carrier. With this choice the impulse imparted to the electron by the electric field of the laser pulse during its duration will be zero. The polarization plane is assumed to coincide with (x, y) plane. The vector potential is taken, in Gaussian units, as

$$A(t,\delta)=-c\int_0^t dt'\,E(t',\delta) \qquad (2)$$

so that it turns to be zero for $t\le 0$ and $t\ge \tau$. By assuming the negative ion initially in a bound state $\psi_i(r, t)$ the transition amplitude for detachment into the final continuum state $\psi_f^-(r,t)$ at the pulse end, in the length gauge, has the form (in atomic units)

$$T_{fi}(\delta)=-i\int_0^\tau dt\left\langle\psi_f^-(r,t)\left|E(t,\delta)\cdot r\right|\psi_i(r,t)\right\rangle \qquad (3)$$

In our calculations $\psi_i(r,t)$ is approximated by

$$\psi_i(r,t)=\psi_i(r)\exp(-iI_0 t) \qquad (4)$$

with

$$\psi_i(r)=A\,r^{-1}\,exp(-kr)Y_{l,m}(\hat{r}) \qquad (5)$$

In Eq. (4) A is the normalization coefficient, $Y_{l,m}(\hat{r})$ a spherical harmonic, and l, m the angular momentum quantum numbers of the electron in the initial state. The quantization axis is chosen along the propagation direction of the laser pulse. $I_0=-k^2/2$ is the energy of the initial bound state. The final state of the detached electron $\psi_f^-(r,t)$ is described by a Volkov state with momentum q . By using the above approximate wavefunctions for $\psi_i(r,t)$ and $\psi_f^-(r,t)$, we have for the transition amplitude at the time t

$$T_{fi}(\delta)=i\int_0^\tau\left\langle\psi_f(r)\left|r\cdot E(t,\delta)\right|\psi_i(r)\right\rangle exp\left\{i\,S_q(t,\delta)\right\}dt \qquad (6)$$

where

$$\psi_f(r)=exp\left\{i\left[q+k_L(t,\delta)\right]\cdot r\right\} \qquad (7a)$$

and

$$S_q(t, \delta) = \int^t dt \left\{ \frac{[q + k_L(t, \delta)]^2}{2} + I_0 \right\} \tag{7b}$$

with $k_L(t,\delta)=A(t,\delta)/c$. Once $T_{fi}(\delta)$ is known, the momentum distribution of the photoelectron in the (x,y) polarization plane is obtained as

$$\frac{d^2 P_{fi}(\delta)}{dq_x \, dq_y} = \int_{-\infty}^{+\infty} |T_{fi}(\delta)|^2 \, dq_z \tag{8}$$

Below, Eq. (8) will be used to calculate the distribution of the photoelectron produced during the detachment of F⁻ ions. The detachment threshold of F⁻ depend on the atomic state in which the atom is left after the process occurs. They are $I_{0\ 1/2}=3.4502\ eV$ and $I_{0\ 3/2}=3.4001\ eV$ for, respectively, the $^2P^0_{1/2}$ and $^2P^0_{3/2}$ atomic final state. The calculation involves summation of photodetachment probabilities for the different values $m=0,\ \pm1$ of the magnetic quantum number characterizing the initial state of the active electron and the statistical averaging of detachment channels associated with the two spin-orbit sublevels $P_{1/2}$ and $P_{3/2}$ of the final atomic states.

3. Results and discussion

Figure 1 shows the photoelectron distribution at the pulse end for various values of the cycle number n_c, putting $E_0=0.0292\ a.u.$, $\omega=0.030\ a.u.$ and $\delta=0$. The photoelectron distributions obtained by Eq.(8) are averaged over the spatial intensity in the laser focus, which is assumed to have a Gaussian form with focal parameters near to those estimated in [11] and $E_0^2/2$ the peak intensity.
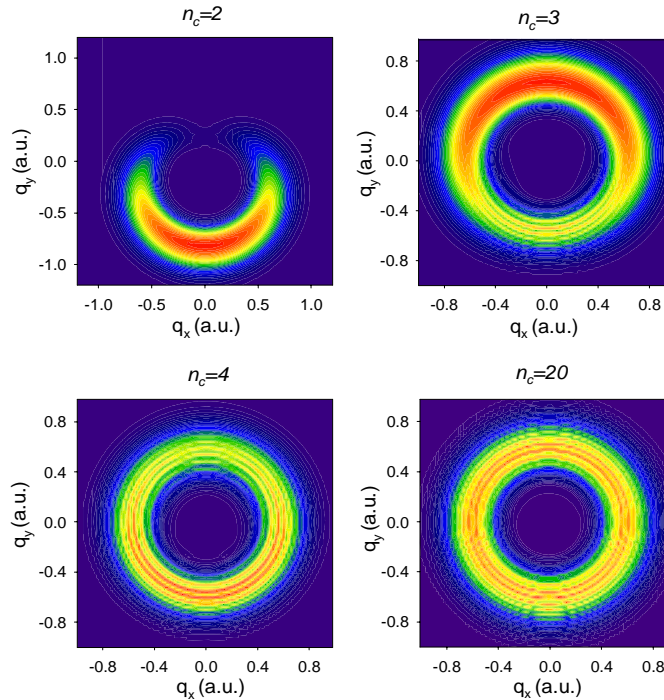

Fig. 1. Averaged electron momentum distribution in the (x,y) polarization plane of the laser pulse for different values of the cycle numbers ($n_c=2, 3, 4, 20$). The photoelectron distribution are averaged over a Gaussian spatial intensity with the focal parameters near to those estimated in [11] with $E^2_0/2$ the peak intensity. $E_0=0.0292$ a.u., $\omega=0.030$ a.u. and $\delta=0$.

By inspection of Fig. 1 we note that the momentum distribution in the polarization plane (x, y) is not isotropic and this anisotropy is strongly reduced when the cycle number is increased. This result may be explained qualitatively by using simple classical arguments based on a semiclassical two-step model [21-24]. In the first step the bound electron is detached instantaneously via tunnelling with zero velocity, at the time t_i in which the final strength is around its maximum value. In the second step, the ejected electron propagates like a free particle under the sole action of pulse. Such an approximation is suitable for the photodetachment process as the asymptotic binding potential has a polarization form $u=-\alpha/2r^4$ where α is the dipole polarizability of the atomic core. According to the above model, the impulse imparted to the electron by the laser field in the time interval τ-t_d is given by

$$\boldsymbol{\pi}(\tau,\delta)-\boldsymbol{\pi}(t_d,\delta)=-\int_{t_d}^{\tau}\boldsymbol{E}(t',\delta)dt' \qquad (9)$$

where $t_d \in [0,\tau]$ is the instant of the detachment and $\boldsymbol{\pi}(t,\delta)$ denotes the electron kinematical momentum at the time t. By assuming $\boldsymbol{\pi}(t_d,\delta) \approx 0$, the electron momentum at the end of the laser pulse turns out to be equal to the impulse given by the laser field:

$$\boldsymbol{\pi}(\tau,\delta)=-\int_{t_d}^{\tau}\boldsymbol{E}(t',\delta)dt'=-\int_{0}^{\tau}\boldsymbol{E}(t',\delta)dt'+\int_{0}^{t_d}\boldsymbol{E}(t',\delta)dt' \qquad (10)$$

As the temporal shape of the laser pulse has been chosen in such a way that the first integral in the rhs of the above equation vanishes, we find that, qualitatively, the electron kinetic momentum at the end of the laser pulse is given by the vector

$$-\int_{0}^{t}\boldsymbol{E}(t',\delta)dt'=1/c\,\boldsymbol{A}(t_d,\delta)=-\boldsymbol{k}_L(t_d,\delta)$$
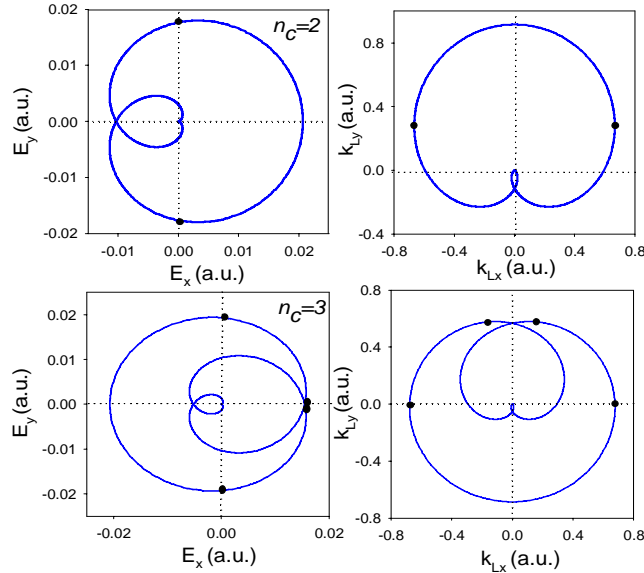
evaluated at the detachment time.

Fig. 2. The left panel curves show the time-dependent electric field $\boldsymbol{E}(t, 0)$ for n_c=2, 3. The corresponding curves for the momentum $\boldsymbol{k}_L(t, 0)$ imparted to the electron by the laser pulse are reported in the right panel. In the left curves the symbols mark the time interval extrema where the electric field is more intense. The same symbols are also marked in the corresponding curves for $\boldsymbol{k}_L(t, 0)$. The laser parameters as in Fig.1.

In Fig. 2, the time-dependent electric field $\boldsymbol{E}(t, 0)$ and the corresponding time-dependent momentum $\boldsymbol{k}_L(t, 0)= -1/c\boldsymbol{A}(t, 0)$ imparted to the electron by the laser pulse in the time interval

[0, t], are respectively shown in the left and right panels, for n_c=2 and 3. For n_c=2, the electric field rotates in the polarization plane describing a curve which, in the time interval [$3\tau/8$, $5\tau/8$], is very close to a semi-circumference of ray approximately equal to its maximum value. In the same time interval $k_L(t, 0)$ rotates describing a quasi-semi-circumference, rotated of $\pi/2$ respect to that described by $E(t, 0)$, lying in the half-plane y>0. Since the kinetic momentum at the end of the pulse is equal to $-k_L(t_d, 0)$, the electron distribution results essentially confined in the half-plane y<0. This in agreement with the plot shown in figure 1 for n_c=2.

The same above considerations can be repeated for n_c=3 in the interval [$5\tau/12$, $7\tau/12$] noting that in this case the vector $k_L(t, 0)$ lies in the half-plane y<0. Consequently, the electrons are preferentially ejected in the half-plane y>0. However, we point out that in the two intervals [$\tau/3$, $5\tau/12$] and [$7\tau/12$, $2\tau/3$] the laser electric field strength is greater than ¾ of its maximum and $k_L(t, 0)$ lies in the half-plane y>0. Therefore, a sizeable electron emission in the plane y<0 may be expected too, as it is shown in Fig.1 for n_c=3.

The plots of Fig. 1, for n_c=4 and 20, may be explained too by considering the time evolution of the vector $E(t, 0)$ and $k_L(t, 0)$. In fact, by increasing n_c, the vectors $E(t, 0)$ and, hence, $k_L(t, 0)$ rotate several times keeping in each turn a quasi constant strength that makes the ejected electron distributions, in the (x,y) plane, almost isotropic. Finally, we remark that above threshold ionization processes caused by the interaction of a few-cycle circularly polarized pulse with hydrogen like atoms have been recently studied by Milosevich et al.[13]. By using the saddle point method they have established the vector potential evaluated at the real part of the saddle points, giving the main contribution to the integral entering the transition amplitude, is almost aligned along the momentum of the ejected electrons. Moreover, by increasing the number of the optical cycles encompassed in a pulse, as a results of the interference effects, well-resolved peaks appear in the curve, evaluated numerically, showing the ionization probability as a function of the energy of the ejected electron. For few-cycle pulse (n_c=2 or 3), it may be happen that, for some angular interval of the ejection direction, the main contribution to the ionization amplitude come from a single saddle point; in this case structureless energy distribution of the photoelectron will be obtained. The result of Ref. [13] applied to the case of photodetachment may be used to explain the curves shown in Fig. 1.
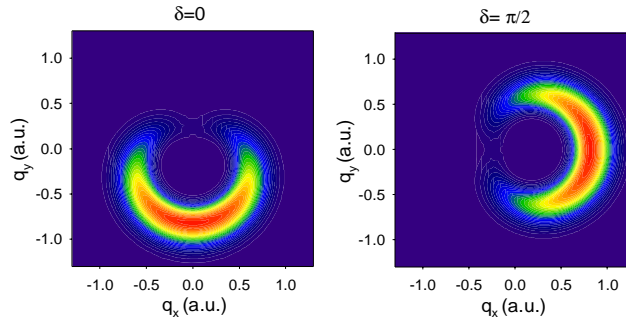

Fig. 3. Averaged electron momentum distribution in the (x,y) polarization plane of the laser pulse for two values of δ with n_c=2. The values of E_0 and ω are the same as in Fig.1.

In Fig. 3 we present the averaged electron momentum distribution in the polarization plane for various values of the carrier-envelope relative phase δ, for n_c=2 and I_L and ω the same as in Fig. 1. By varying the carrier-envelope relative phase from 0 to δ, the electron momentum distribution rotates counter clock wise of an angle equal to δ. Then we may conclude that a change in the carrier-envelope relative phase from 0 to δ corresponds to a rotation of our system around the z-axis of the angle φ=δ. This occurs since the transition amplitude $T_{fi}(\delta)$, Eq. (2), is equal to that evaluated at δ=0 in a system rotated by an angle -δ around the z-axis

$$T_{fi}(\delta) = -i\int_0^\tau dt \left\langle \psi_f(\boldsymbol{r},t) \middle| \boldsymbol{E}(t,\delta)\cdot\boldsymbol{r} \middle| \psi_i(r,t) \right\rangle$$

$$= -i\int_0^\tau dt \left\langle \psi_f(\boldsymbol{r},t) \middle| T^+(\delta)T(\delta)\boldsymbol{E}(t,\delta)\cdot\boldsymbol{r}\, T^+(\delta)T(\delta) \middle| \psi_i(r,t) \right\rangle \qquad (11)$$

$$= -i\int_0^\tau dt \left\langle \psi_f(\boldsymbol{r}',t) \middle| \boldsymbol{E}(t,0)\cdot\boldsymbol{r} \middle| \psi_i(\boldsymbol{r}',t) \right\rangle$$

In Eq. (10) $T(\delta)$ is the unitary operator corresponding to a rotation of our system by an angle -δ around the z-axis and $\boldsymbol{r}' = T(\delta)\,\boldsymbol{r}$.
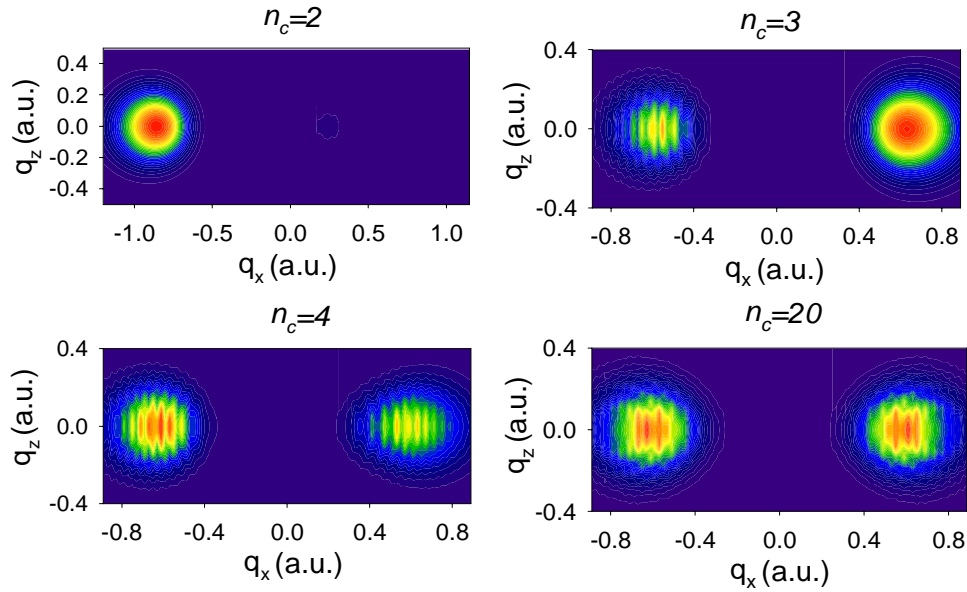

Fig.4 Averaged electron momentum distribution in the (x,z) plane, perpendicular to the polarization plane of the laser pulse, for different values of the cycle number (n_c=2, 3, 4, 20). The laser parameters as in Fig.1

Figure 4 shows the averaged electron momentum distribution in the (y,z) plane. In agreement with the classical picture, few electrons are emitted along the z-axis. The plots exhibit asymmetries around the line q_y=0, which are strongly reduced when the cycle number increases. For n_c=20 the asymmetries, in agreement with the electron momentum distribution recorded in ref.[11], disappear. However, as it will be discussed more deeply elsewhere, we remark that the modulations in the momentum distribution shown in Fig. 4 are practically washed out by increasing the spatial laser inhomogeneity.

By summarizing, we have studied the atom threshold detachment of F⁻ by describing the initial bound state in a very simple way. The obtained results have been analyzed in terms of a two-step semiclassical model. Accordingly, after the electrons are detached via tunnelling, their motion is determined by the action of the electric field pulse following the classical dynamics laws. Anisotropies in the angular distribution of the electrons ejected on the plane perpendicular to the laser field propagation direction are found that decrease by increasing the number of cycles of the laser pulse.

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