CHIN. PHYS. LETT. Vol. 34, No. 6 (2017) 063201

Angle-Resolved Electron Spectra of F^- Ions by Few-Cycle Laser Pulses *

Jian-Hong Chen(陈建宏)[1**], Song-Feng Zhao(赵松峰)[2], Guo-Li Wang(王国利)[2], Xiao-Ping Zheng(郑小平)[1], Zheng-Rong Zhang(张正荣)[1]

[1]*School of Electronic and Information Engineering, Lanzhou City University, Lanzhou 730070*
[2]*College of Physics and Electronic Engineering, Northwest Normal University, Lanzhou 730070*

(Received 3 March 2017)

The above-threshold detachment of F^- ions induced by a linearly polarized few-cycle laser pulse is investigated theoretically using the strong-field approximation model without considering the rescattering mechanism. We first derive an analytical form of transition amplitude for describing the strong-field photodetachment of F^- ions. The integration over time in transition amplitude can be performed using the numerical integration method or the saddle-point (SP) method of Shearer et al. [Phys. Rev. A 88 (2013) 033415]. The validity of the SP method is carefully examined by comparing the energy spectra and photoelectron angular distributions (PADs) with those obtained from the numerical integration method. By considering the volume effect of a focused laser beam, both the energy spectra and the low-energy PADs calculated by the numerical integration method agree very well with the experimental results.

PACS: 32.80.Gc, 32.80.Qk, 32.80.Rm DOI: 10.1088/0256-307X/34/6/063201

With the rapid advancements in electron imaging spectroscopy (EIS) and high-resolution recoil-ion momentum spectroscopy (RIMS) techniques,[1,2] the angle-resolved electron spectra from negative ions have become subjects of great interest in strong-field physics.[3–24] Experimentally, Kiyan *et al.* investigated the energy- and angle-resolved electron spectra for H^- ions[2–4] and F^- ions,[5–8] respectively. In particular, an unusual maximum in the photoelectron angular distributions (PADs) of H^- ions was reported experimentally[2] and then well reproduced by using the strong-field approximation (SFA) model and solving the time-dependent Schrödinger equation (TDSE).[9] The saddle-point (SP) method was used successfully to understand the unusual maximum in the PADs of H^- ions in our previous work.[22]

The SP method based on the KFR-type theory[10] has been widely used to study the laser-induced photodetachment of negative ions. Recall that the original SP method was developed by Gribakin and Kuchiev under adiabatic approximation.[10] That is to say, the original SP method is valid only when the frequency of the laser field ω is much lower than the electron binding energy I_p. However, the condition of adiabatic approximation is not suitable for most current intense field experiments. Fortunately, recent works[25,26] based on the SP method have shown a clear nonadiabatic effect in the tunneling region with the Kelysh parameter $\gamma \sim 1$, which suggests that the SP method still works well as an effective tool for studying strong-field effects. Moreover, with an infinitely long periodic laser pulse, the SP method was used to interpret the experimental angle-resolved photoelectron spectra by considering the volume effect. Recently, to achieve the optical control of laser-induced photodetachment of negative ions, Shearer *et al.* have extended the SP method to the case of few-cycle linearly polarized laser pulses.[17,18] However, $2(N+1)$ exact solutions of saddle points must be obtained numerically for N-cycle laser pulse in the generalized SP method[17–22] and thus it is rather time-consuming if the volume effect of laser pulses is included. To bypass the time-consuming difficulty of the generalized SP method, in this Letter, we first derive the above-threshold detachment (ATD) probability amplitude formula for F^- ions exposed to an intense laser field. It is found that the numerical calculations based on our formula take much less time than those performed by the generalized SP method. The validity of the SP method is examined carefully by comparing with the energy spectra, the momentum spectra and the PADs from numerical integration method based on our formula for a single peak intensity. When the volume effect of the laser beam is included, it is found that the angle-resolved energy spectra using the SFA are in good agreement with the experimental results. Atomic units are used in this study unless otherwise stated.

In the first-order SFA, i.e., the SFA model without considering the rescattering mechanism, the direct transition probability amplitude from the atomic or ionic ground state $|\psi_0\rangle$ to the continuum state $|\psi_{\boldsymbol{p}}\rangle$ with momentum $\boldsymbol{p}$ is given by[8,9]

$$A_{\boldsymbol{p}} = -i \int_{-\infty}^{+\infty} \langle \psi_{\boldsymbol{p}} | H_i(t) | \psi_0 \rangle dt, \tag{1}$$

where $H_i(t) = \boldsymbol{r} \cdot \boldsymbol{E}(t)$ is the electron–laser interaction in the length gauge, and $\boldsymbol{E}(t)$ is the electric field of the laser pulse. The initial state of F^- ions can be

*Supported by the National Natural Science Foundation of China under Grant Nos 11647150, 11464026, 11664035 and 11364038, the Young Talents Program of Gansu Province in 2016, the Scientific Research Program of the Higher Education Institutions of Gansu Province under Grant No 2016A-068, and the Doctoral Scientific Research Foundation of Lanzhou City University under Grant No LZCU-BS2015-04.

**Corresponding author. Email: chenyuwen1982@163.com

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approximated as an asymptotic form[18]

$$\psi_0(\boldsymbol{r}) = \frac{A}{r}\exp(-\kappa r)Y_{10}(\hat{r}), \tag{2}$$

where $\kappa = \sqrt{2I_\mathrm{p}}$, $I_\mathrm{p} = 3.401\,\mathrm{eV}$ is the ionization potential of F^- ions, and $A = 0.7$ represents an asymptotic constant for the wave function. The final state is approximately by the Volkov state

$$\psi_{\boldsymbol{p}}(\boldsymbol{r},t) = \exp\Big[i(\boldsymbol{p}+\boldsymbol{A}(t))\cdot\boldsymbol{r} - \frac{i}{2}\int_{-\infty}^{t}(\boldsymbol{p}+\boldsymbol{A}(t'))^2dt'\Big]. \tag{3}$$

The vector potential of the laser pulse is defined by $\boldsymbol{A}(t) = -\int^t \boldsymbol{E}(t')dt'$. The electric field of a linearly polarized laser pulse (along the z-axis) can be written as

$$\boldsymbol{E}(t) = E_0\cos^2\left(\frac{\pi t}{\tau}\right)\cos(\omega t)\hat{z}, \tag{4}$$

for the time interval $(-\tau/2,\tau/2)$ and zero elsewhere, and E_0, ω and τ are the amplitude, angular frequency and total duration of the laser pulse, respectively.

After performing the integration over space coordinates in Eq. (1) analytically, the transition probability amplitude for F^- ions can be rewritten as

$$A_{\boldsymbol{p}} = -i\frac{\sqrt{6}}{2\pi}A\int_{-\infty}^{+\infty}dt\exp\Big(-\frac{i}{2}\int_{-\infty}^{t}q^2dt' + iI_\mathrm{p}t\Big)E(t)\times\Big[\frac{q^2-3q_z^2}{q^5}\arctan\frac{q}{\kappa} + \frac{5q^2q_z^2\kappa+3q_z^2\kappa^3-q^4\kappa-q^2\kappa^3}{q^4(q^2+\kappa^2)^2}\Big], \tag{5}$$

where $\boldsymbol{q} = \boldsymbol{p}+\boldsymbol{A}(t')$ is a canonical momentum of the electron in a laser field, and q_z is the projection of $\boldsymbol{q}$ in the polarization direction of the laser pulse.

In Eq. (5), the transition amplitude $A_{\boldsymbol{p}}$ is usually calculated by numerically integrating over time (hereafter to be called the numerical integration method). Alternatively, $A_{\boldsymbol{p}}$ can also be evaluated using the SP method of Shearer *et al.*[18,19] as follows:

$$A_{\boldsymbol{p}} = -(2\pi)^{3/2}A\sum_{t_\mathrm{s}}(\pm1)^l Y_{lm}(\hat{q}_\mathrm{s})\frac{\exp(if(t_\mathrm{s}))}{\sqrt{-if''(t_\mathrm{s})}}, \tag{6}$$

where $\hat{q}_\mathrm{s}$ is the unit vector in the direction of the complex canonical momentum $\boldsymbol{p}+\boldsymbol{A}(t_\mathrm{s})$, and the sum runs over all SPs t_s which are the complex roots of the following equations in the upper half-plane,

$$(\boldsymbol{p}+\boldsymbol{A}(t_\mathrm{s}))^2 + 2I_\mathrm{p} = 0. \tag{7}$$

In practice, exact saddle points of complex t_s can be obtained by solving Eq. (7) with the Newton–Raphson method[17–19] or the Secant method.[20–22] In Eq. (6), $f(t_\mathrm{s})$ and $f''(t_\mathrm{s})$ are given by

$$f(t_\mathrm{s}) = -\int_{t_\mathrm{s}}^{\infty}\Big[\frac{1}{2}(\boldsymbol{p}+\boldsymbol{A}(t'))^2 + I_\mathrm{p}\Big]dt', \tag{8}$$

$$f''(t_\mathrm{s}) = -2\boldsymbol{p}\cdot\boldsymbol{E}(t_\mathrm{s}) - 2\boldsymbol{A}(t_\mathrm{s})\cdot\boldsymbol{E}(t_\mathrm{s}). \tag{9}$$

In this study, we choose the orbital quantum number $l = 1$ and the magnetic quantum number $m = 0$ for F^- ions, since this electron state gives a dominant contribution to the detachment signal.[19]

The momentum distribution of the photoelectrons is given by

$$\frac{\partial^2 P}{\partial E\partial\theta} = |A_{\boldsymbol{p}}|^2 2\pi p. \tag{10}$$

Due to the cylindrical symmetry of the system in a linearly polarized laser field, the energy spectra of the detached photoelectrons can be obtained by

$$\frac{\partial P}{\partial E} = \int\frac{\partial^2 P}{\partial E\partial\theta}\sin\theta d\theta, \tag{11}$$

where θ is the angle between the emission direction of the ejected photoelectron and the polarization axis of the laser field.

To compare with the experimental results, one has to consider the volume effect of a focused laser beam. We assume that the gas volume is larger than the laser-focus volume in this work. Thus the ejected electron signals are the sum of detached electrons within the interaction volume. For a peak intensity I_peak, the yield of the photoelectrons with momentum $\boldsymbol{p}$ is given by[27,28]

$$S(\boldsymbol{p},I_\mathrm{peak}) = \Gamma\int_0^{I_\mathrm{peak}}|A_{\boldsymbol{p}}(I)|^2\Big(-\frac{\partial V}{\partial I}\Big)dI, \tag{12}$$

where Γ is the density of target atoms in the interaction volume, $A_{\boldsymbol{p}}(I)$ denotes the transition amplitude for a single intensity I, and $-\frac{\partial V}{\partial I}dI$ represents the volume element between the laser intensities I and $I+dI$. We assume that the spatial distribution of the laser intensity is Lorentzian in the propagation direction z and Gaussian in the transverse direction. Therefore the volume element for this laser beam is given as[27]

$$-\frac{\partial V}{\partial I}dI = \frac{\pi W_0^2 x_\mathrm{R}}{3}\frac{1}{I}\Big(\frac{I_\mathrm{peak}}{I}+2\Big)\sqrt{\frac{I_\mathrm{peak}}{I}-1}dI, \tag{13}$$

where W_0 is the $1/e$ radius of the focal spot, and x_R is the Rayleigh range of the focus. Without loss of generality, we assume $W_0 = x_\mathrm{R} = \Gamma = 1\,\mathrm{a.u.}$ in our simulations.

When the volume effect is considered, the momentum distribution of the photoelectrons is rewritten as

$$\frac{\partial^2 S}{\partial E\partial\theta} = |S(\boldsymbol{p},I_\mathrm{peak})|^2 2\pi p. \tag{14}$$

The angle-resolved electron spectra of F^- ions exposed to a 100 fs linearly polarized laser pulse have been reported in Ref. [7]. To simulate the experimental data, we take a three-cycle laser pulse with wavelength of 1400 nm and peak intensity of $1.7\times10^{13}\,\mathrm{W/cm^2}$ shown

in Fig. 1(a). For this laser pulse, the ponderomotive energy $U_p = 3.12$ eV and the Keldysh parameter $\gamma = 0.74$, thus the electron detachment is well in the tunneling regime.

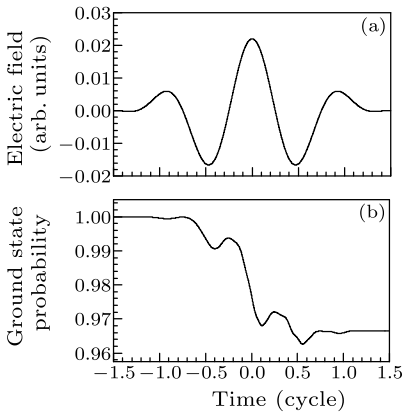

Fig. 1. (a) The electric field of a three-cycle laser pulse with wavelength of 1400 nm and peak intensity of 1.7×10^{13} W/cm^2. (b) Time evolution of the ground-state survival probabilities for F^- ions in the laser field shown in Fig. 1(a).

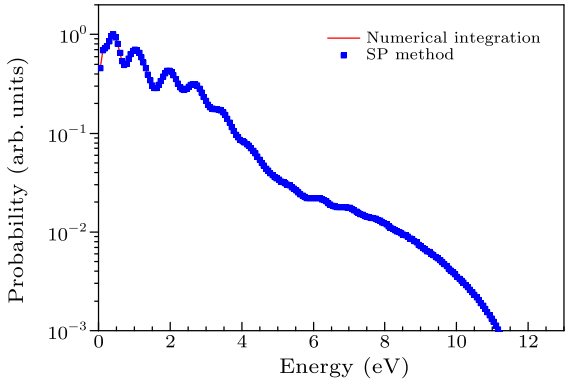

Fig. 2. The normalized energy spectra of laser-induced photoelectron of F^- ions in a three-cycle laser pulse with wavelength of 1400 nm and peak intensity of 1.7×10^{13} W/cm^2.

In Fig. 1(b) we show the ground-state survival probability of F^- ions in a three-cycle laser pulse calculated by solving the time-dependent Schrödinger equation (TDSE) with the generalized pseudospectral technique.[29–31] In our simulations, the converged TDSE results can be obtained by setting $r_{max} = 200$, the maximum radial grid point $N_r = 400$, the time step $dt = 0.1$ and the maximum partial waves $l_{max} = 40$. For F^- ions, we use the double Yukawa potential,[9] i.e., $V(r) = -a_1 e^{-a_2 r}/r - a_3 e^{-a_4 r}/r$, where a_1, a_2, a_3 and a_4 are 5.137, 1.288, 3.863, and 3.545, respectively. One can see that the largest depletion of ground-state probabilities is less than 4% for the three-cycle short laser pulse as shown in Fig. 1(b). Therefore, we will ignore the depletion of ground-state population in the following SFA calculations.

In this study, we present the detailed probability amplitude formula of F^- ions (see Eq. (5)) based on the first-order SFA model. The SP method will be examined carefully by comparing the energy spectra, the momentum spectra and the photoelectron distributions with those obtained from the numerical integration based on Eq. (5). Figure 2 shows the energy spectra of laser-induced photoelectrons of the F^- ions. For simplicity, all the energy spectra are normalized to 1.0 at the first peak. It can be seen that the energy spectra from these two methods agree perfectly with each other. In Fig. 3, we present the two-dimensional (2D) momentum spectra of the photoelectrons of the F^- ions. Here $p_\perp$ and $p_{||}$ represent the momentum components perpendicular and parallel to the polarization direction of the laser's electric field, respectively. Note that the momentum spectra obtained from the numerical integration and from the SP method are normalized to 1.0 at the maximum of detachment probabilities and the color is plotted on the logarithmic scale. One can see that the structures of momentum spectra from the numerical integration method and from the SP method agree very well with each other. To more directly compare the results from these two methods, Fig. 4 shows the PADs from these two methods for photoelectron energies at 1.6 eV, 3.4 eV, and 6.7 eV, respectively. All the PADs are normalized to 1.0 at the peak. It can be seen that the PADs calculated from these two methods are almost the same as expected.

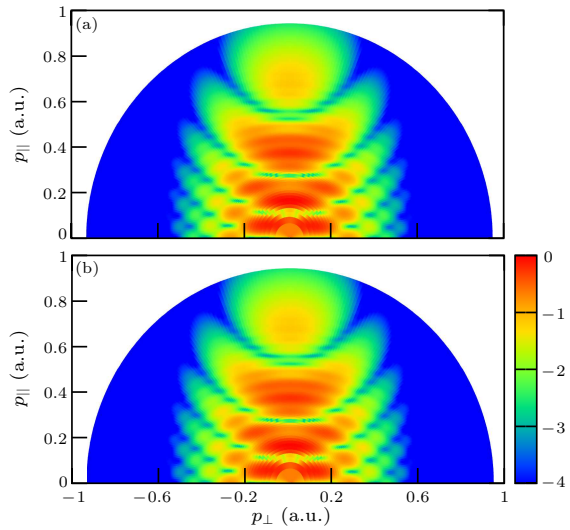

Fig. 3. The normalized momentum spectra of the photoelectrons detached from F^- ions in a three-cycle laser pulse by using the (a) numerical integration method and (b) the saddle-point method. The color is plotted on the logarithmic scale. The laser parameters are the same as Fig. 2.

In the following we investigate the effect of the spatial inhomogeneity of a focused laser beam (i.e., the volume effect) on the photoelectron energy spectra and PADs of the F^- ions. In Fig. 5 we compare the experimental energy spectra and PADs with those theoretical results obtained from the numerical integration method by including the volume effect. It should be mentioned that all the energy spectra and PADs are normalized to 1.0 at the peak. Figure 5(a) shows the energy spectra of laser-induced photoelectron of F^- ions at the ejection angles $\theta = 0°$ and $\theta = 90°$, respectively. It is found that the agreement

between the experimental energy spectra and the theoretical ones is quite good in general. In Figs. 5(b)–5(d), we compare the PADs for photoelectron energies at 1.6 eV, 3.4 eV and 6.7 eV, respectively. It can be seen that the volume-integrated PADs from the numerical method can reproduce reasonably the main features of the experimental data. It is found that there are large differences in the single-intensity based PADs and the volume-integrated PADs by comparing Fig. 4 with Fig. 5, especially for lower energies such as 1.6 eV and 3.4 eV. It is indicated that the volume effect of the laser beam is very important for quantitatively reproducing the experimental results of the ATD of F^- ions in an intense laser field.

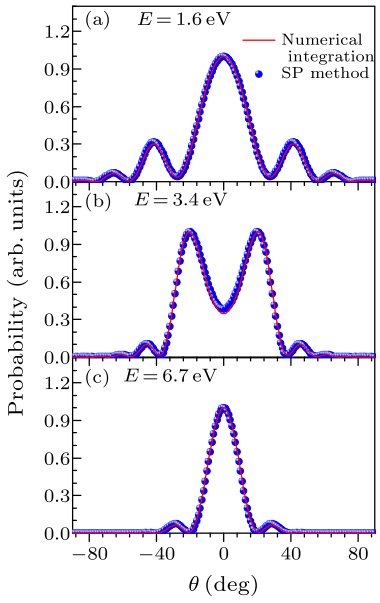

Fig. 4. The normalized photoelectron angular distributions of F^- ions for (a) the electron energy $E = 1.6$ eV, (b) $E = 3.4$ eV and (c) $E = 6.7$ eV. The laser parameters are the same as Fig. 1(a).

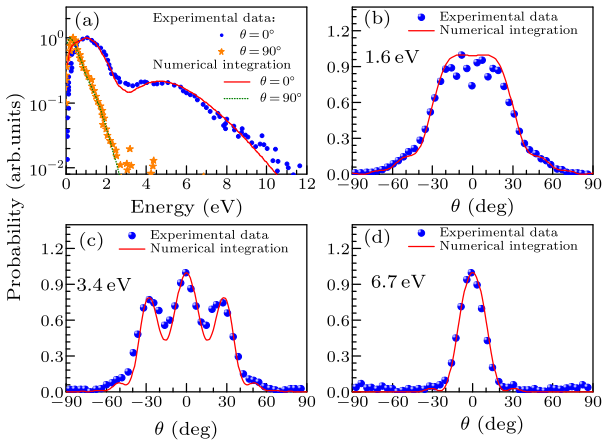

Fig. 5. (a) The normalized energy spectra of laser-induced photoelectron of F^- ions at $\theta = 0°$ and $\theta = 90°$. The normalized photoelectron angular distributions of F^- ions for (b) the electron energy $E = 1.6$ eV, (c) $E = 3.4$ eV and (d) $E = 6.7$ eV. The volume effect is included in our numerical integration calculations to compare with the experimental data from Bergues *et al.*[7] The laser parameters are the same as Fig. 1(a).

In summary, the direct photo-detachment of electrons from F^- ions by a three-cycle linearly polarized laser pulse has been investigated theoretically. Based on the first-order SFA, we derive the formula of probability amplitude which describes the strong-field photodetachment dynamics of F^- ions. The validity of the SP method has been examined by comparing with the corresponding energy spectra, momentum spectra and the PADs obtained from the numerical integration method based on Eq. (5). Finally, we calculate the photoelectron energy spectra at two fixed angles and PADs of the F^- ions by including the volume effect of the laser beam and then compare with the experimental data. We find that the first-order SFA model only considering the volume effect can be used to reproduce the experimental low-energy photoelectron spectra and PADs of F^- ions in an intense laser field. The methods will be extended to study the direct ATD of complex negative ions by ultrashort laser pulses in the future.

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