Experimental Study of Photodetachment in a Strong Laser Field of Circular Polarization

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Negative fluorine ions are exposed to a circularly polarized infrared laser pulse with a peak intensity on the order of 2.6×10^{13} W/cm^2. A fundamental difference, as compared to the case of linearly polarized field, is found in the absence of any structure in the photoelectron spectrum that can be associated with the quantum interference effect. This observation is in accord with our recent predictions [S. Beiser *et al.*, Phys. Rev. A **70**, 011402 (2004)]. The experiment reveals that the length gauge is appropriate for the description of the field interaction in the frame of the strong field approximation.

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With the development of powerful lasers, the elementary process of ionization in a strong electromagnetic field received great attention. From the theoretical side, numerous nonperturbative approaches have been developed to describe this effect. One of them, the Keldysh-Faisal-Reiss (KFR) theory [1–3], has the advantage of treating the problem analytically. The KFR theory uses an approximation that consists of neglecting the binding potential in the final state. Such an approximation is suitable for the description of photodetachment of an electron bound by a short-range potential. Atoms do not belong to this category of atomic systems because of the long-range Coulomb interaction of the detached electron with the residual core. In negative ions, in contrast, the asymptotic binding potential has a polarization form $U = -\alpha/(2r^4)$, where α is the dipole polarizability of the atomic core. This potential vanishes much faster than the Coulomb potential at large distances r. Therefore, negative ions represent a suitable atomic system for testing the KFR theory.

The KFR theory has been developed further in several publications by using a saddle point analysis of the transition amplitude [4,5]. This analysis turned out to be successful in explaining the origin of modulations that appear in spectra of photoelectrons detached in a *linearly* polarized laser field. These modulations are well pronounced in angle resolved energy spectra of photoelectrons produced in the direct process of photodetachment [4] as well as in the process that involves electron rescattering on the parent core [5]. The theory describes this effect as being due to interference of quantum paths leading to the same final momentum state of the outgoing electron. This interference effect was already observed in photodetachment of negative ions by linearly polarized field [6,7]. In the limit of low electron momentum, the interference pattern reduces to that of a two slit interference [8].

The question whether the interference effect occurs in photodetachment by a circularly polarized laser field has been addressed theoretically in Refs. [9,10]. The saddle point analysis of the transition amplitude reveals only one saddle point contribution for circular polarization, indicating the absence of interference in this case. One goal of the present work is to verify this result experimentally.

Both the velocity and the length gauge were used in the KFR theory for the description of the electron-field interaction. The discrepancy between predictions by different gauges was discussed for the case of circularly polarized field in Ref. [10]. It was shown that in the length gauge the transition amplitude depends on both absolute value and sign of the magnetic quantum number m of the initial state. In contrast, in the velocity gauge it depends only on the absolute value of m. It has also been shown that only predictions by the length gauge are consistent with the Wigner threshold law [11] in the limit of low electron momentum. This fact indicates that the length gauge is the proper one for the KFR theory. A similar conclusion is also given for the case of linear polarization in Ref. [12], where results of the numerical solution of the time-dependent Schrödinger equation are compared with predictions of the KFR theory. Another goal of the present work is to solve the gauge controversy experimentally. Below we compare our measurements with predictions in both gauges.

In the present work we study photodetachment of F^- in a strong laser field of circular polarization. As in our previous experiments [6], the difficulty of exposing negative ions to high laser intensities is overcome by using an infrared laser pulse of 100 fs duration. In such a short pulse the electron acceleration due to the gradient of the intensity distribution in the laser focus can be neglected [13]. The experimental apparatus is basically the same as used before [14]. Negative fluorine ions are extracted from a hollow cathode glow discharge operated with a gas mixture of CF_4 and krypton. The ions are accelerated to a kinetic energy of 3 kV, mass selected in a Wien filter and focused to a waist of 0.4 mm size in an ultrahigh vacuum chamber. A typical ion current of F^- is 100 nA, as measured with a Faraday cup after the interaction region. Differential pumping allows us to maintain the interaction chamber at 10^{-10} mbar during the experiment.

Infrared laser pulses of 1.510 μm wavelength are generated in an optical parametric amplifier (OPA) pumped with a mode-locked Ti:sapphire laser system at a 1 kHz repetition rate. The output of OPA has linear polarization. It is converted to circular polarization by using an achromatic quarter wave plate. We characterize the degree of polarization by a parameter ϵ, which we introduce by describing the field strength as $\mathbf{F}(t) = F(\cos(\omega t), (1 - \epsilon)\sin(\omega t), 0)$. Here the polarization plane is assumed to be the xy plane. We measured the ellipticity parameter ϵ by rotating a Wollaston prism polarizer after the wave plate and measuring the intensities of the two beam components as a function of the rotation angle. The ellipticity is found to be less than 1%, which is the precision limit of this measurement. For such a high degree of circular polarization, the orientation of the ellipse in the interaction chamber can be disregarded in the data analysis, and we do not discuss it further. The laser beam is focused with a 15 cm focal length lens into the interaction region. It crosses the ion beam at 90° and propagates parallel to the detector plane. The focus size of 53 μm (FWHM) and the pulse length of 100 fs (FWHM) are obtained with the use of our homebuilt tools for beam diagnostic. Using these parameters and assuming a Gaussian shape of the spatial and temporal pulse profiles, the peak intensity in the interaction region is found to be 2.6×10^{13} W/cm^2. The Keldysh parameter at these experimental conditions is approximately 0.8.

An electron imaging spectrometer operated in the velocity mapping regime [14] is used to detect the photodetached electrons. The image processing involves a conventional Abel inversion routine [15]. The Abel inversion requires the electron distribution to be axially symmetric with its symmetry axis parallel to the detector plane. This condition is satisfied in the present case, since in the circularly polarized field electrons are ejected symmetrically with respect to the laser propagation direction. The momentum calibration of the detector was taken from our previous measurement [6].

Figure 1 shows the momentum distribution of background-subtracted photoelectrons obtained after Abel inversion of the raw data. Here $p_{\parallel}$ and $p_{\perp}$ represent the momentum components parallel and perpendicular to the polarization plane, respectively. Because of the ponderomotive broadening, the excess photon detachment channels are not resolved here. The ponderomotive energy shift corresponding to the peak intensity in the focus is $U_p = 5.5$ eV, while the photon energy is 0.82 eV. In the polarization plane the spectrum extends up to momenta of 1.3 a.u. corresponding to a kinetic energy of approximately 23 eV, and it involves absorption of more than 28 excess photons. The minimum number of photons required to overcome the detachment threshold of F^- at the given wavelength is 5. In contrast to the case of photodetachment in a linearly polarized laser pulse, the spectrum is smooth

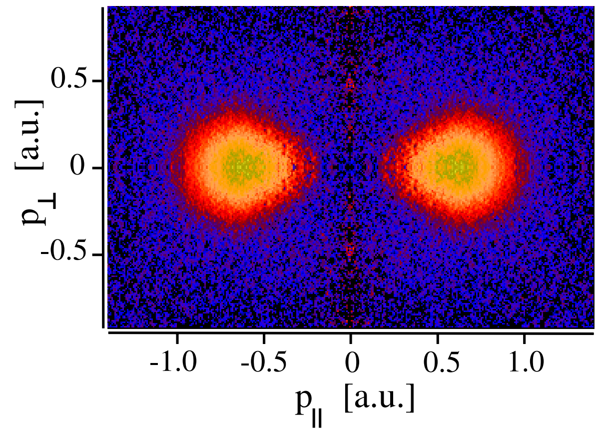

FIG. 1 (color online). Momentum distribution of photoelectrons. $p_{\parallel}$ and $p_{\perp}$ represent the momentum components parallel and perpendicular to the plane of circular polarization, respectively.

and does not exhibit any nonmonotonic features indicating a quantum interference effect. In agreement with the simple classical picture, electrons are preferentially detached in the polarization plane while no electrons are emitted along the laser propagation axis.

We simulated photoelectron spectra in the length and velocity gauge by using corresponding analytical expressions for the detachment rate given in Refs. [3,10], respectively. The simulation procedure is the same as described in Ref. [7]. It involves summation of contributions to the detachment rate from the initial state with different values of the magnetic quantum number $m = 0, \pm 1$, as well as integration of the electron yield over the laser focus with the measured spatiotemporal intensity distribution. Finally, the spectra are convoluted with the response function of the detector.

Simulations in both gauges produce a smooth momentum distribution of photoelectrons that resembles the experimental distribution presented in Fig. 1. In both cases the electron spectrum appears monotonic with its maximum in the polarization plane. For a quantitative comparison, we plot in Fig. 2 the experimental energy distribution of photoelectrons in the polarization plane together with the predicted spectra. The upper and lower parts of the figure show a comparison with simulation results in the velocity and length gauge, respectively. Theoretical and experimental data are normalized to each other according to the averaged signal in the vicinity of its peak value. The dashed lines represent predictions performed at the measured peak intensity of 2.6×10^{13} W/cm^2, whereas the full lines show predictions at a peak intensity of 3.8×10^{13} W/cm^2, which is approximately 45% higher than the measured value. We did already note in our previous experiment with linearly polarized light that a much better agreement between the experiment and the KFR theory is obtained when in the simulation the peak intensity is allowed to be higher [7]. This fact has also been pointed

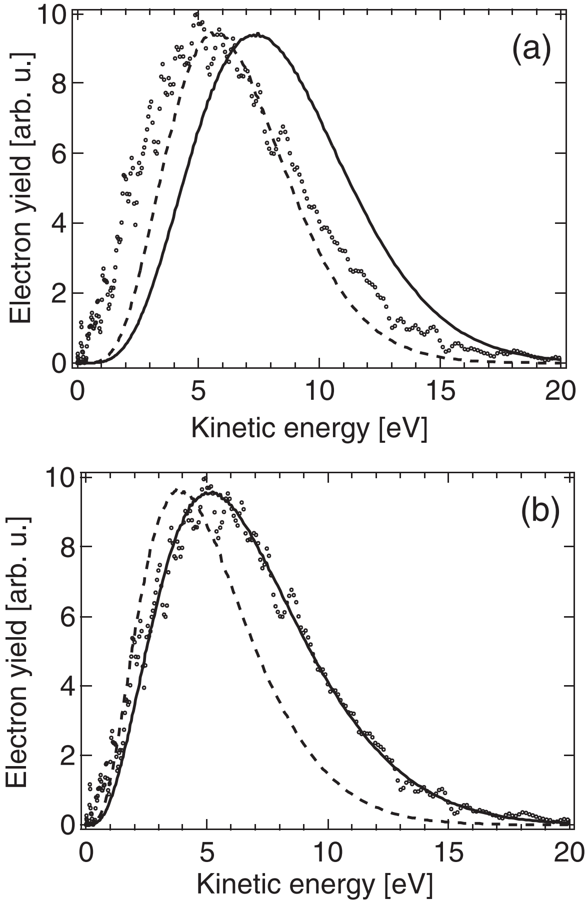

FIG. 2. Experimental energy distribution of photoelectrons emitted in the polarization plane (dots) compared with theoretical predictions in the velocity gauge (a) and length gauge (b). Dashed lines show simulation results for the measured peak intensity of 2.6×10^{13} W/cm^2. Solid lines represent predictions for a 45% higher peak intensity of 3.8×10^{13} W/cm^2.

out in a previous comparison of theory and experiment presented in Refs. [16,17], where a 20% and a 60% higher intensity, respectively, was needed to reproduce our experimental results. We estimate the precision of the peak intensity measurement to be on the order of 15%. This value does not include possible systematic errors, such as a pointing instability of the laser beam during the focus size measurement and a deviation of the temporal and spatial intensity distribution from a Gaussian shape. For example, a pointing instability of 0.2 mrad would lead to the overestimation of the focus size resulting in the underestimation of the peak intensity by 45%. Nevertheless, Fig. 2(b) shows a very good agreement between the experimental data and predictions in the length gauge, if one allows for a 45% higher peak intensity. In contrast, predictions in the velocity gauge fail to reproduce the measured data. Even if the peak intensity is taken as an "adjustable parameter," the energy spectrum in the velocity gauge is still too narrow to fit the experimental data. This picture has been totally reproduced in our second experiment (not presented here) performed at a slightly higher peak intensity of 3.2×10^{13} W/cm^2. In the second measurement we also observe a perfect agreement between experimental data and predictions in the length gauge, when the simulation is performed for a 50% higher peak intensity than the measured value.

Figure 3 shows the angular distribution of electrons at a kinetic energy of 5.1 eV, corresponding to the maximum of the energy spectrum in the polarization plane. Here θ denotes the angle with respect to the polarization plane. The measured results are compared with the two predicted results normalized to the maximum of experimental data. Simulations are shown for the measured peak intensity. We found that for a given kinetic energy the angular distributions predicted from theory are largely insensitive to the value of the peak intensity, and theoretical curves in Fig. 3 appear the same when this value is 45% higher. One can see from the figure that the length gauge predicts a slightly broader distribution than the velocity gauge. The comparison with the experiment is in favor of the length gauge, especially on the wings of the angular distribution. This fact is not restricted to the chosen kinetic energy of 5.1 eV: comparison of angular distributions at lower and higher kinetic energies (not shown here) yield similar results.

As already discussed in Ref. [10], a tremendous discrepancy between predictions in the length and velocity gauges appear when the initial bound state has a nonzero angular momentum and one needs to take into account photodetachment from the initial state components characterized by different magnetic quantum numbers m. Consider the present case of F^- where the angular momentum of the outer electron is 1. While in the velocity gauge contributions from the initial state components with $m = -1$ and $m = +1$ are identical, predictions in the length gauge reveal a huge difference in their contributions to the photo-

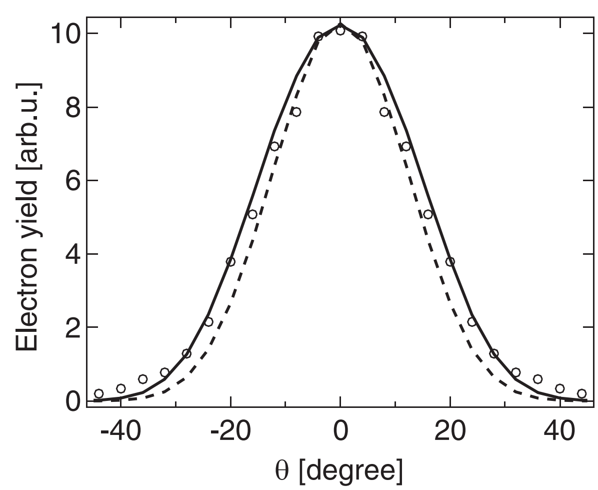

FIG. 3. Angular distribution of electrons with a kinetic energy of 5.1 eV. Circles represent experimental data; solid and dashed lines show predictions in the length and velocity gauge, respectively.

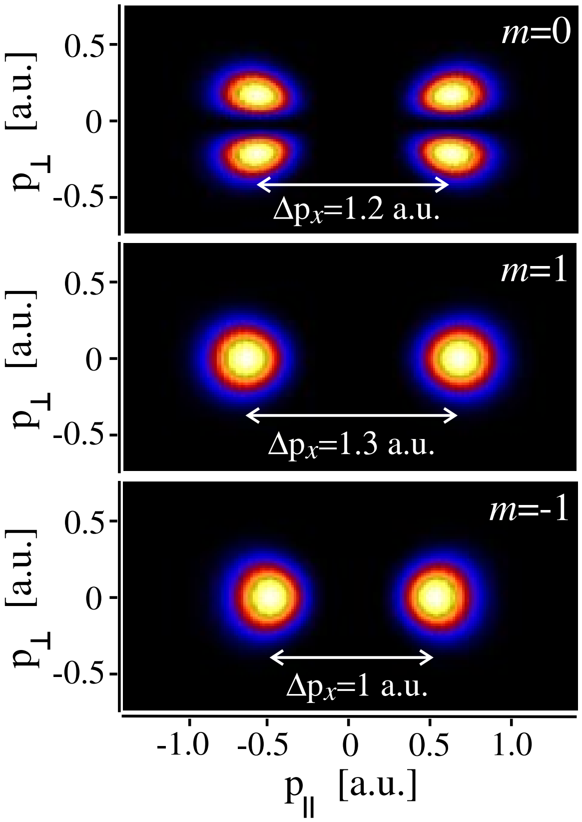

FIG. 4 (color online). Simulations of the momentum distribution for different m components of the initial state. The maxima of the $m = 0$ and $m = 1$ contributions are lower by a factor of 23 and 3, respectively, compared to the maximum of the $m = -1$ contribution.

electron spectrum. It was also pointed out in Ref. [10] that only predictions in the length gauge can reproduce the Wigner threshold law of photodetachment, which is also being due to the m dependency of the photodetachment rate. In order to illustrate this matter, we present in Fig. 4 simulated momentum distributions of photoelectrons in the $(p_\|, p_\perp)$ coordinates separately for each m component of the initial state. Calculations are performed in the length gauge for the measured peak intensity and wavelength. It should be noticed that for a visual convenience the three distributions are normalized to the same maximum value of the electron yield, so that they can easily be compared on the same color scale. The maximum values of calculated results are, actually, quite different. The highest contribution appears from the $m = -1$ component, which is approximately 3 times higher than the $m = 1$ contribution. The lowest electron yield is provided by the $m = 0$ component, with its maximum value approximately 23 times lower than for the $m = -1$ contribution. The sum of these three distributions is to be compared with the distribution shown in Fig. 1, and it does resemble the experimental spectrum. We point out the difference between the $m = -1$ and $m = 1$ contributions, which is evident from Fig. 4. Despite the similarity in their shape, one can see a well pronounced shift of their maxima relative to each other. On the energy scale this shift is 2.3 eV, corresponding to approximately three photon energies. Such a dramatic difference in the energy position of the maximum of electron distribution can be well resolved experimentally, once a suitable candidate ion with alignment is prepared for a photodetachment study.

In conclusion, we have proven experimentally that the quantum interference effect, which governs the process of photodetachment in a linearly polarized laser field, is absent in the case of circular polarization. The comparison of experimental data with predictions by the KFR theory reveals that the length gauge is proper to use in the frame of the strong field approximation. It remains challenging to develop this study to the case where the initial state is prepared with a particular nonzero magnetic quantum number m, and to demonstrate the dependency of the photoelectron spectrum on the sign of m.

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