Electron Rescattering in Above-Threshold Photodetachment of Negative Ions

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We present experimental and theoretical results on photodetachment of Br^- and F^- in a strong infrared laser field. The observed photoelectron spectra of Br^- exhibit a high-energy plateau along the laser polarization direction, which is identified as being due to the rescattering effect. The shape and the extension of the plateau is found to be influenced by the depletion of negative ions during the interaction with the laser pulse. Our findings represent the first observation of electron rescattering in above-threshold photodetachment of an atomic system with a short-range potential.

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The rescattering of a photoelectron on its parent core represents a fundamental effect that lies in the focus of recent investigations in the field of the nonlinear interaction of atoms and molecules with strong laser radiation. An electron liberated by photoionization moves in the presence of the laser field. Driven by a field of linear polarization, it may revisit the parent core. Its recollision with the core gives rise to processes such as high-order above-threshold ionization (HATI), high-order harmonic generation, and nonsequential multiple ionization (see the review articles [1,2] and references therein).

The HATI process, which represents the subject of the present work, manifests itself in photoelectron spectra as a high-energy plateau stretching along the laser polarization axis. On the energy scale this plateau extends up to (and has a cutoff at) approximately $10U_p$, where U_p is the electron ponderomotive energy in the laser field. The HATI process was found to be initiated by elastic rescattering of the photoelectron on its parent core, where the highest kinetic energy is reached via backscattering [3]. More recently, the HATI process has received much attention in the context of attophysics [4], as a means to measure the carrier-envelope phase of a few-cycle laser pulse in a single shot [5], and also since it allows one to extract the differential cross sections for elastic electron scattering off positive ions from the experimental angle-resolved photoelectron spectra [6].

While the rescattering effect has been widely studied in atoms and molecules, its role in negatively charged ions remains essentially unexplored [7]. One might expect the manifestation of rescattering in above-threshold photodetachment of negative ions to be different. The short-range character of binding forces in negative ions precludes the Coulomb focusing of the electron wave packet created in the continuum. Coulomb focusing was shown to enhance the rescattering probability [8]. On the other hand, the lower binding energy of negative ions implies a larger initial size of the electron wave packet and, consequently, its slower spreading between the instants of ionization and rescattering. Whether or not these features result in a significant modification of the HATI process represents a fundamental question. In the present work we report on the first observation of the rescattering plateau in above-threshold photodetachment of negative ions.

Early experiments on strong-field photodetachment were hampered by depletion of the negative-ion sample at the leading edge of the laser pulse. This problem was overcome by using a short laser pulse of infrared wavelength and, thus, by reaching the saturation condition of photodetachment at a significantly higher intensity. First experiments with infrared pulses of 100 fs duration were performed by our group on H^- [9] and F^- [10]. The peak intensity was of the order of 10^{13} W/cm^2. Though electrons of rather high kinetic energies were detected in [10], the dynamic range of the signal, limited to 2.5 orders of magnitude, did not allow us to distinguish a rescattering contribution from the experimental background. The results were described well by using a Keldysh-like theory [11] that considers the direct detachment only.

In the present work we investigate photodetachment of Br^-. Our study is motivated by the recent prediction that the ratio of the rescattering signal to the signal of direct electrons is higher for the heavier halogen negative ions, Br^- and I^- [12]. Intuitively, heavier elements have a larger core, which gives rise to a larger elastic scattering cross section. Recently, we reported on photodetachment of Br^- in a laser field of 800 nm wavelength and 6×10^{14} W/cm^2 peak intensity [13]. A jet of energetic electrons along the laser polarization axis, resembling a rescattering plateau, was observed in the measured spectra. This jet, however, was found to be due to sequential double detachment. If there was any yield from the rescattering process, it was

masked by the dominant double-detachment signal. In the present work the contribution of sequential double detachment is suppressed by exposing Br^- to radiation of a longer wavelength of 1300 nm. At this wavelength the nonlinearity of the double-detachment process is much higher and the signal arising from sequential double detachment lies far below the noise level of the measured spectra.

Our experimental setup is described elsewhere [9,10,13]. Briefly, a mass-selected beam of negative ions is intersected with the laser beam inside an electron imaging spectrometer (EIS) operated in the velocity mapping regime. Linearly polarized infrared laser pulses of 1300 nm wavelength are generated in an optical parametric amplifier (OPA) pumped with a mode-locked Ti:sapphire laser system at a repetition rate of 1 kHz. The output of the OPA is focused with a 15 cm focal length lens into the interaction region. A focus size of 40 μm (FWHM) and a pulse duration of 100 fs (FWHM) are measured with the use of our beam diagnostic tools. Assuming a Gaussian shape of the spatiotemporal intensity distribution, the peak intensity in the focus is determined to be 6.5×10^{13} W/cm^2. The image processing involves a conventional Abel inversion routine to reconstruct the angle-resolved momentum distribution of the photoelectrons emitted from the laser focus. The momentum/energy resolution of the EIS is determined by measuring the detector response function to a single electron event. The energy resolution is 0.23 eV at the energy of 1 eV, and it varies proportionally to the square root of the kinetic energy.

We analyze the measured spectra in terms of the rescattering theory developed in Ref. [12]. This rescattering theory is an extension of the Keldysh-Faisal-Reiss theory [14] and yields a first-order correction to the strong-field approximation (SFA). In the standard SFA, the interaction of the photoelectron with the residual core is neglected. This approximation is particularly suitable for the description of photodetachment of negative ions due to the absence of the long-range Coulomb potential, which is experienced by the outer electron in atoms. However, even for negative ions a photoelectron, driven back into the inner region of the binding potential, may interact with the atomic core. The rescattering theory of Ref. [12] allows for such an interaction. A closely related approach was formulated in Ref. [15] and successfully applied to photodetachment of F^- [16].

Let us stress the main aspects of this rescattering theory. The probability amplitude to detach an electron with a drift momentum $\mathbf{p}$ is defined by the matrix element [2]

$$M_{\mathbf{p}i} = -i \lim_{t\to\infty} \int_{-\infty}^{t} dt' \langle \psi_{\mathbf{p}}(t) | U(t,t') \mathbf{r}\cdot\mathbf{E}(t') | \psi_i(t') \rangle, \quad (1)$$

where $U(t,t')$ is the time-evolution operator of the Hamiltonian $H(t) = -\nabla^2/2 + \mathbf{r}\cdot\mathbf{E}(t) + V(\mathbf{r})$, $\mathbf{r}\cdot\mathbf{E}(t)$ is the laser-field–electron interaction given in the length gauge and dipole approximation, and $V(\mathbf{r})$ is the electron-atom interaction in the absence of the laser field. The wave functions $\psi_{\mathbf{p}}$ and ψ_i describe the final state with the drift momentum $\mathbf{p}$ and the initial state, respectively. As discussed in detail in Ref. [11], ψ_i can be represented in the asymptotic form $\psi_{\ell m}(\mathbf{r}) = (A/r)\exp(-\kappa r) Y_{\ell m}(\hat{\mathbf{r}})$, where A is a normalization constant, $E_a = \kappa^2/2$ is the binding energy, and ℓ, m are the angular-momentum quantum numbers of the initial state. The time-evolution operator $U(t,t')$ satisfies the Dyson equation

$$U(t,t') = U_L(t,t') - i \int_{t'}^{t} dt'' U_L(t,t'') V(\mathbf{r}) U(t'',t'), \quad (2)$$

where $U_L(t,t')$ is the time-evolution operator of the Hamiltonian $H_L(t) = -\nabla^2/2 + \mathbf{r}\cdot\mathbf{E}(t)$ of a free electron in the laser field. We now make two approximations: on the right-hand side of Eq. (2) we replace U with U_L, and in Eq. (1) we replace the final state $\langle\psi_{\mathbf{p}}(t)|$ by a plane wave. The state $\langle\psi_{\mathbf{p}}(t)|U_L(t,t')$ then becomes the Volkov state $\langle\psi_{\mathbf{p}}^{(L)}(t')|$ (eigenstate of a free electron in the external laser field), and we obtain

$$M_{\mathbf{p}i}^{\mathrm{SFA}} = -i \int_{-\infty}^{\infty} dt \langle \psi_{\mathbf{p}}^{(L)}(t) | \mathbf{r}\cdot\mathbf{E}(t) | \psi_i(t) \rangle - \int_{-\infty}^{\infty} dt \int_{t}^{\infty} dt' \langle \psi_{\mathbf{p}}^{(L)}(t') | V U_L(t',t) \mathbf{r}\cdot\mathbf{E}(t) | \psi_i(t) \rangle. \quad (3)$$

The first term on the right-hand side of Eq. (3) represents the probability amplitude of direct detachment (the standard SFA), while the second term describes the (first-order Born approximation) rescattering amplitude. It is the second term that gives rise to the high-energy plateau in the electron energy spectrum [12]. For above-threshold detachment off a negative ion—in contrast to above-threshold ionization of an atom—we expect Eq. (3) to yield a quantitatively reliable description.

The rescattering amplitude is dependent on $V(\mathbf{r})$, which is specific for a given negative ion. The potential $V(\mathbf{r})$ can be modeled by the double Yukawa potential [17]

$$V(r) = -\frac{Z}{H}\frac{e^{-r/D}}{r}[1 + (H-1)e^{-Hr/D}]. \quad (4)$$

Here $H = DZ^{0.4}$, Z is the atomic number and D is a numerical parameter. For bromine $Z = 35$, $D = 0.684$ and, for comparison, for fluorine $Z = 9$, $D = 0.575$ [18]. The potential (4) has a static character and does not include polarization effects. It was shown in [12] that these effects are not significant for electron rescattering at high kinetic energies. Therefore, for the sake of simplicity we use the static potential and demonstrate below to which extent the difference in the parameters Z and D for Br^- and F^- affects the rescattering signal from these negative ions.

For a fixed laser intensity I and frequency ω, the energy spectrum of photoelectrons consists of a series of discrete peaks at energies $E_{\mathbf{p}} \equiv \mathbf{p}^2/2 = n\omega - E_a - U_p$ with $n \geq n_{\min}$, where $U_p = I/(4\omega^2)$ and $n_{\min}$ denotes

the minimum number of photons needed to overcome the ponderomotively shifted detachment threshold. For a detailed comparison with the experimental results, the electron spectrum has to be averaged over the spatiotemporal intensity distribution in the laser focus. Since the intensity in our experiment is far above the saturation value, the depletion of the negative ions during the interaction with the laser pulse also needs to be taken into account [9,10]. Details on the simulation routine will be presented elsewhere. The experimental energy resolution is taken into account by convolution of the simulated spectra with the measured response function of the detector to a single electron event.

Our experimental results and the predictions by the rescattering theory are presented in the upper and lower part of Fig. 1, respectively. The figure shows the measured and calculated photoelectron distributions in the (p, θ) coordinates, where p is the electron momentum and θ is the emission angle with respect to the laser polarization axis. Despite the limited signal-to-noise ratio at low signal and the presence of background in the experimental spectrum, Fig. 1 demonstrates good agreement between experiment and theory.

In the following discussion we focus on electron detachment along the laser polarization axis, where the rescattering contribution is most pronounced. The measured and simulated kinetic-energy spectra of electrons emitted at $\theta = 0°$ are presented in Fig. 2. Here we also show results of simulations where rescattering is not taken into account. These are obtained by disregarding the second term in Eq. (3) that describes the rescattering amplitude. Comparison of the results in Fig. 2 clearly demonstrates that rescattering contributes to the yield of electrons with kinetic energies higher than 10 eV. At this energy the experimental curve changes its slope and starts deviating considerably from the predictions for the direct electrons, while it is well reproduced by the predictions with rescattering taken into account. The rescattering signal dominates in the spectrum at energies higher than 15 eV and appears well pronounced in the range up to approximately 35 eV. Beyond this value it becomes indistinguishable from the experimental background. In the intermediate range between 10 and 15 eV both the direct-detachment and the rescattering amplitudes in Eq. (3) are significant and interfere. In this region the slope changes gradually to a smaller value, which characterizes the "plateau."

The upper energy limit of the observed spectrum, however, is much lower than the cutoff energy of $10U_p$, which has a value of 103 eV for the measured peak intensity. The signal of rescattered electrons continuously decreases with increasing energy, forming an inclined plane rather than a plateau. Such an unusual manifestation of the rescattering plateau is found to be due to strong saturation of the photodetachment process. Indeed, by tracing in our simulations the population density of negative ions during the interaction with the laser pulse, we obtain that only 1% of the ions in the laser focus survive until the intensity reaches the value of 3×10^{13} W/cm^2, which is still a factor of 2 lower than the measured peak intensity. In other words, only a very small number of negative ions are practically exposed to the peak intensity. Integration of the product of the population density and the photodetachment rate over the focal region results in the inclined-plane shape of the rescattering signal without a pronounced cutoff energy. It follows from this discussion that the condition of strong saturation makes the peak intensity less crucial. This has also been verified by performing simulations for different peak intensities that differ from the measured value by up to 30%, which is our estimate for the intensity error bar.

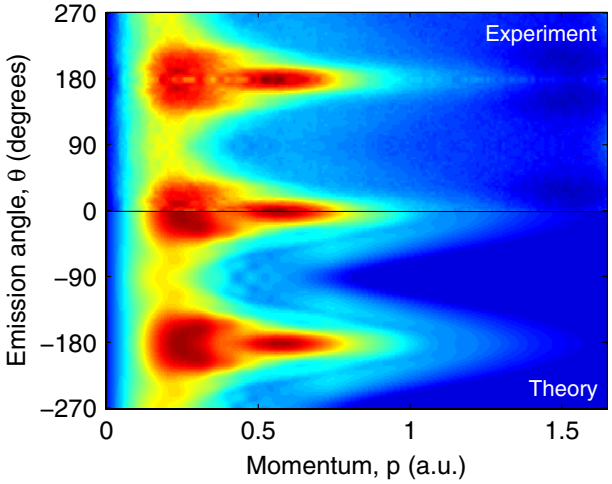

FIG. 1 (color online). Angle-resolved momentum distribution of photoelectrons detached from Br^- in a laser field of 1300 nm wavelength and 6.5×10^{13} W/cm^2 peak intensity. Upper part ($0° \leq \theta \leq 270°$): experimental results; lower part ($-270° \leq \theta \leq 0°$): predictions by the rescattering theory.

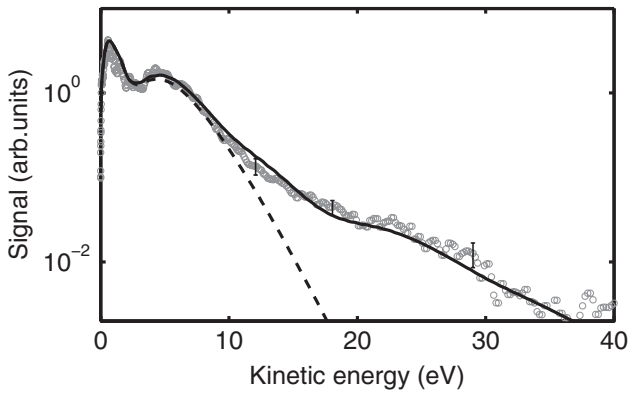

FIG. 2. Electron energy distributions along the laser polarization axis obtained from the spectra shown in Fig. 1. Circles: experiment; solid line: predictions with rescattering taken into account. The dashed line shows the predictions for the direct electrons only. The theoretical distributions are normalized to the experimental data at the maximum of the signal. Error bars of a few experimental data points are shown.

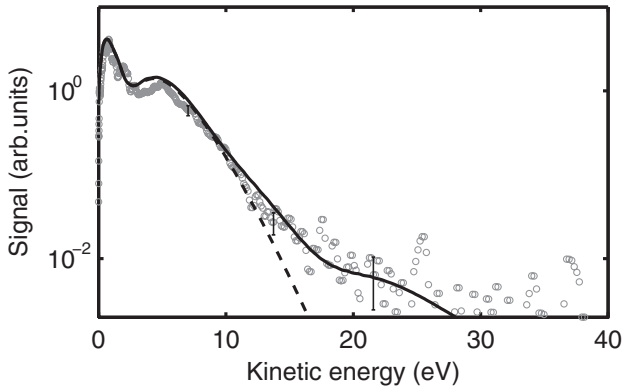

FIG. 3. Same as Fig. 2 but for F^-. The laser wavelength is 1300 nm and the peak intensity is 3.4×10^{13} W/cm^2.

The variation of the peak intensity does not cause significant changes in the predicted spectra.

Let us consider, for comparison, photodetachment of F^- whose rescattering potential (4) is described by different parameters Z and D. The smaller values of the parameters Z (smaller potential depth) and D (steeper decrease of the potential with the increase of r) reflect the smaller size of the fluorine atom as compared to the bromine atom. Results on photodetachment of F^- in a strong infrared laser field have been presented before [10]. Here we present new data obtained for F^- at the wavelength of 1300 nm, as used in the experiment on Br^-. Because of a different optics alignment (focus size of 45 μm, pulse duration of 133 fs), the value of the peak intensity in this measurement was 3.4×10^{13} W/cm^2. Figure 3 shows measured and calculated energy distributions of electrons emitted along the laser polarization axis. The two predicted distributions are obtained with and without taking rescattering into account. Only the onset of the rescattering signal in the energy range between approximately 15 and 22 eV can be distinguished from the noise in the measured spectrum. Except for this range, the predicted rescattering plateau, which again looks more like an inclined plane, lies below the experimental background. One should note that the dynamic range of the signal is the same in both F^- and Br^- data and is restricted to approximately 3 orders of magnitude. This prevents us from studying the rescattering effect in F^-. The smaller contribution of the rescattering effect in F^- as compared to Br^- is a consequence of the smaller size of the fluorine core. This is in accord with the predictions of Ref. [12].

In conclusion, we reported on the first observation of the HATI process in negative ions. The shape of the rescattering signal is found to be strongly affected by saturation of the photodetachment process. Because of saturation, the rescattering plateau assumes the form of an inclined plane. This fact reflects the fragility of negative ions compared to atoms and positive ions. Because of a larger core, the rescattering contribution to the spectrum is larger for heavier elements. This makes observation of rescattering effects easier in Br^- than in F^-.

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