Coulomb Asymmetry in Above-Threshold Ionization

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A new method for including effects of the Coulomb potential in strong-field laser atom interaction is presented. The model is tested by comparing its results with experimental data of energy resolved angular distributions of photoelectrons. For elliptical polarization these exhibit a strong asymmetry. Our theory shows that this strong asymmetry for the low-energy electrons is induced by a small Coulomb force acting on the tunneling electron just after the exit of the tunnel. This is in contrast to the situation for high electron energies where the asymmetry arises via rescattering by the parent ion.

Recent developments in strong-field laser physics—in particular, the unfolding field of attosecond physics—has generated new interest in the fascinating phenomena observed there. It is characteristic for the field that many features measured for harmonic generation, single photoionization, and multiple photoionization are rather similar for different atoms, ions, and molecules. Such a remarkable similarity of different quantum systems suggests that the observed properties are governed more by the laser field rather than by the structure of the atomic species (typically rare gas atoms).

This tendency was divined by Keldysh's nonperturbative approach to strong-field photoionization [1], which is now widely used and known as strong-field approximation (SFA). In this approach, the effect of the laser field on the electron motion after ionization is treated exactly while the effect of the binding potential is neglected. The Keldysh approach bypasses a difficult problem: While the Schrödinger equation for an electron in either a Coulomb or a laser field can be solved analytically, the combination of both fields is very challenging. This holds, in particular, for intense laser radiation, where laser and Coulomb field are of comparable strength.

Notwithstanding the important and beautiful insights provided by the SFA, it has been clear for years that the neglect of the atomic potential can cause severely wrong results. The necessity to revisit this long-standing problem has been underlined by new experimental and theoretical results obtained for multiple ionization (for a review see [2]) and for extremely short laser pulses [3]. Here we address the problem of the Coulomb field in strong-field photoionization. At high intensity ($I > 5 \times 10^{13}$ W/cm$^2$) atoms may absorb more photons than necessary for ionization (above-threshold ionization, ATI). In the photoelectron spectra this is seen in an electron yield extending to energies much higher than the photon energy; for review see Refs. [4,5].

The first striking evidence for Coulomb effects in strong-field laser atom interaction has been found in ATI spectra generated with elliptical polarization. The theory developed along the lines of SFA by Perelomov, Popov, and Terent’ev (the PPT theory) predicts a fourfold symmetry of the photoelectron angular distributions with respect to reflections about both main axes of the polarization ellipse [6]. In contrast, the experimentally observed distributions [7] possess only inversion symmetry; i.e., they are asymmetric in any half of the polarization plane. A number of theoretical calculations beyond the SFA [8–10] as well as a general analysis of the symmetry properties of the multiphoton ionization amplitude for atoms [11] and negative ions [12] have led to the conclusion that the fourfold symmetry is a direct consequence of the core assumption of the SFA approach entirely neglecting the influence of the binding potential on the electron motion in the continuum. So far, however, all attempts to achieve reasonable agreement with theoretical models for ionization of neutral atoms failed. For negative ions no detectable asymmetry was observed [13].

Other earlier work [14] has concentrated on the angular distributions of the so-called plateau electrons. These high-energy ($> 25$ eV) electrons are generated through a rescattering process during ionization. A generalization of the SFA that includes rescattering delivers results in reasonable agreement with the experiment. From such an analysis it can be seen that the shape of the angular distributions of the high-energy (i.e., plateau) electrons depends mostly on the kinematics of the electron motion between the instants of ionization and rescattering [15] and much less on the shape of the scattering potential. The data in the low-energy part of the spectra were not analyzed due to the lack of a relevant theory [14].

Here we present new high-resolution experimental data on ATI with elliptical laser polarization that show unprecedented details and are a good benchmark to study the Coulomb effects. For the reason given above we concentrate on low-energy (or “direct”) electrons, i.e., electrons that did not rescatter. The goal of this paper is to develop a theory capable to describe the data and to elucidate the mechanism of Coulomb effects in this part of the spectrum. Actually, we formulate a surprisingly appealing semiclassical model which is based on the SFA...
approach and incorporates the effect of the Coulomb field of the ion on the ionization process. It is well known that strong-field ionization can be viewed in two separate steps: tunneling out of an atom and classical motion in the laser field along the orbit to a detector [16,17]. This provides the possibility to introduce Coulomb corrections for each step separately. The effect of the ion field on tunneling was considered earlier for static [18] and for low-frequency [19] electric fields. Although the latter correction increases the total ionization rate by several orders in magnitude, it does not affect the fourfold symmetry of the angular distributions [see the factor $C(F)$ in Eq. (1) below]. In our new model, we consider the impact of the Coulomb field on the electron motion after ionization. Moreover, under the present experimental conditions (high intensity and intermediate ellipticities not too close to zero) the effect of the ion's Coulomb field can be evaluated perturbatively along the electron trajectory in the laser field. It will turn out that just this small correction produces dramatic changes in the photoelectron distributions and destroys the fourfold symmetry.

Consider an atom with binding energy $I$ subject to the elliptically polarized field $\mathbf{F}(t) = F(\sin \omega t, \xi \cos \omega t, 0)$ with ellipticity $\xi$. In short laser pulses, energy conservation allows ionization to quantum states with drift momentum $\mathbf{p}$ satisfying the condition $p^2/2 = (N + s)\omega - I - F^2(1 + \xi^2)/4\omega^2$, where $N$ is the minimum number of absorbed photons necessary to reach continuum and $s = 0, 1, 2, \ldots$ is a number of ATI peak. Under the conditions $\omega \ll 1$ and $F \ll F_a = (2I)^{3/2}$, the corresponding amplitude of ionization can be calculated in the framework of the SFA approximation. ($F_a$ is the atomic electric field at the first Bohr orbit in a hydrogen-like atom with the ionization potential $I$. We use atomic units, i.e. $e = m = \hbar = 1$.) Evaluation of the integral determining this amplitude with the saddle-point method allows for to substantiate the two-step description of direct ionization [20]. In brief, in the tunneling regime, $\gamma = \sqrt{2I}/F \ll 1$, and with interferences being ignored [21], a contribution to the differential ionization rate from a single saddle-point is of the form:

$$\frac{dW}{dt_0 \, d\nu_0 \, dp_z} = \frac{C^2(F(t_0)) \omega}{4\pi^2 \sqrt{\varepsilon_p(t_0) [1 + \varepsilon_p(t_0)/I]}} \exp \left[-\frac{2F_a^2}{3\sqrt{\varepsilon_p(t_0)}} \right] \times \left[1 + \frac{\varepsilon_p(t_0)}{I}\right]^{1/2},$$

where $\varepsilon_p(t) = p + A(t)/c$ and $\varepsilon_p(t) = v_p^2(t)/2$ are, respectively, the time-dependent velocity and kinetic energy of an electron moving in the laser field with drift momentum $p$; the factor $C(F) = 2\sqrt{2}F_a/F$ is the Coulomb correction for sub-barrier motion [19]; $t_0 = t_0(p)$ is the real part of the saddle-point $t$. The latter is to be determined from the equation $I + \varepsilon_p(t) = 0$ which, relying upon the condition $\gamma \ll 1$, can be reduced to an equation for $t_0$ of the following form [20]:

$$\mathbf{F}(t_0) \cdot v_p(t_0) = 0.$$  \hspace{1cm} (2)

Below we will discuss the distribution in the polarization plane, i.e., for $p_z = 0$. Then, the velocity in (2) may be written as

$$v_p(t_0) \equiv v_0 = v_0 \mathbf{n}(t_0).$$  \hspace{1cm} (3)

where $\mathbf{n}(t_0)$ is a unit vector in the polarization plane orthogonal to the field $\mathbf{F}(t_0)$ and $v_0$ is an arbitrary parameter. It follows from the definition of the velocity in the laser field that

$$p_x = \frac{F}{\omega} \cos \omega t_0 + v_0 s_x(t_0), \quad p_y = -\frac{F}{\omega} \sin \omega t_0 + v_0 s_y(t_0).$$  \hspace{1cm} (4)

A two-step description is introduced by considering $t_0$ and $v_0$ as new independent variables instead of $p_x$ and $p_y$. They can be interpreted as the time of ionization and the electron velocity at that instant, respectively. By calculating the Jacobian from Eq. (4), one brings the differential rate (1) into the form of a simple-man-type distribution in the time of ionization $t_0$ and in the transverse initial velocity $v_0$:

$$\frac{dW}{dt_0 \, dv_0 \, dp_z} = 0$$

$$= \frac{C^2(F(t_0)) \omega}{4\pi^2 \sqrt{1 + v_0^2/F^2(t_0)}} \times \exp \left[-\frac{2F_a^2}{3\sqrt{F^2(t_0) + v_0^2/F^2(t_0)}} \left[1 + \frac{v_0^2}{2F^2(t_0)}\right]^{1/2}\right].$$  \hspace{1cm} (5)

In Eqs. (5) and (1) we do not assume that the initial velocity is small, $v_0^2 \ll 1$, and, therefore, these equations are applicable for arbitrary values of $v_0$ and $p$, respectively. This is in contrast to the earlier results [6,20] which are valid near the maximum of the momentum distribution only.

If the distribution in initial parameters (5) is adopted to describe strong-field ionization, then one has to calculate the final electron momentum $p$ outside the field, i.e., at $t \to +\infty$. The result of the SFA approach given by Eq. (1) is obtained by integrating Newton’s equation in the laser field which is adiabatically turned off at $t \to +\infty$, with the initial condition for the velocity being specified by Eq. (3). Integration of the time-dependent velocity leads to the electron trajectory in the laser field, $r_z(t, t_0, v_0)$. It is of importance for the following analysis that the starting point of the trajectory $r(t_0) = r_0$ is at the turning point of the classical motion (i.e., at the exit of the tunnel),
The final momentum can be expressed as a sum:

$$r_0 = (1/F(t_0)) n_{F(t_0)}$$

where $n_{F(t_0)} = F(t_0)/F(t_0)$. In the laser field, all solutions can easily be found analytically.

In our model, we also use the distribution of initial conditions (5). However, we calculate the electron motion after ionization, taking into account both laser and ion fields. The direct way to find the final momentum is to integrate Newton's equation (including laser and Coulomb forces) numerically. This was done, for example, in Ref. [23] for the investigation of the Coulomb focusing effect in linearly polarized fields. We shall restrict our consideration to sufficiently large ellipticities. Therefore, the electron trajectory will not come very close to the ion for the vast majority of initial parameters $(t_0, v_0)$. Under these conditions, the Coulomb force is small in comparison to the laser field at the starting point (i.e., at the exit of the tunnel) and decreases further along the trajectory [24]. That allows treating the Coulomb field as a perturbation and evaluating its contribution to the final momentum by integrating the Coulomb force along a trajectory governed by the laser field only:

$$p_c(t_0, v_0) = -\int_0^{t_0} dt \frac{r_c(t, t_0, v_0)}{r_l(t, t_0, v_0)}.$$  \(6\)

Using a subscript $L$ to denote the momentum gained in the laser field [given in Eq. (4)], we write down the actual final momentum as a sum:

$$p = p_L(t_0, v_0) + p_c(t_0, v_0).$$  \(7\)

In order to obtain the Coulomb-affected photoelectron momentum distribution, one chooses an appropriate grid of the initial values $(t_0, v_0)$ and evaluates the probability given by Eq. (5) and the respective final momentum given by Eq. (7). It should be noted that there may be several essentially different initial sets $(t_0, v_0)$ leading to the same final momentum $p$ [25]. Strictly speaking, in such a situation the corresponding transition amplitudes must be added coherently giving rise to interference patterns in the ATI distributions. This effect was observed experimentally with elliptically polarized field [22]. As mentioned before, here we ignore such interference effects and just sum the probabilities incoherently.

Since the Coulomb force drops fairly rapidly with increasing distance from the ion, the Coulomb momentum $p_c$ is mainly gained on the very initial part of the electron trajectory adjacent to the exit of the tunnel. A very good estimate for the Coulomb correction can be obtained by calculating the integral (6) along a straight trajectory generated by the constant field $F(t_0)$. For small initial velocities, $v_0 \ll \sqrt{2I}$, the main contribution is given by

$$p_c(t_0, v_0) = -\frac{\pi \tau_0}{4r_0^2} n_{F(t_0)}.$$

where $\tau_0 = \sqrt{2r_0/F(t_0)}$ is the effective time interval during which the distance from the ion doubles. It happens to be equal to the time of flight under the potential barrier and, hence, is a small fraction of the laser period.

The experimental data were obtained with 40 fs laser pulses. The intensity was varied around $10^{14}$ W/cm$^2$ and different values of the ellipticity were used: $\xi = 0.18$, 0.36, and 0.56. More experimental details can be found in Ref. [14]. Data have been taken for all rare gas atoms except for helium. The energy resolved angular distributions of the photoelectrons in the polarization plane were recorded for 144 azimuthal angles between 0 and $2\pi$. For intensities around $10^{14}$ W/cm$^2$ the low-energy part of the spectrum produced by direct electrons lies below 12 eV. In

![FIG. 1. Polar diagrams of angular distributions of ATI in xenon at an intensity of $\sim 0.9 \times 10^{14}$ W/cm$^2$. The major axis of the polarization ellipse is horizontal. Experimental data are plotted by a thick solid line, the results of the standard Keldysh approximation calculated from (1) are given by a thin solid line, and the results of the present calculations based on Eqs. (6) and (5) are indicated by a dashed line. The left and right panels correspond to ellipticities $\xi = 0.36$ and $\xi = 0.56$, respectively.](image-url)
Fig. 1 the angular distributions recorded for low-energy ATI peaks are presented for the case of ionization of xenon at two different values of the ellipticity. All distributions clearly demonstrate the loss of fourfold symmetry. The position of the maximum in angular distributions virtually does not depend on the order \( s \) of the peak. However, with increasing ellipticity, the maximum approaches the minor axis of the polarization ellipse. The width of the distribution decreases with increasing the peak order \( s \). Lowest peaks with \( s = 1, 2, 3 \) have two maxima of comparable magnitude. For peaks of highest order the second maximum vanishes rapidly.

The results of calculations based on the above described model are also shown in Fig. 1. It can be seen that the agreement with the experimental data, including the position of the maximum and the width of distribution, is quite good except for the first three ATI peaks. The calculations do not reproduce the double hump structure which is clearly seen on the experimental data. We attribute this disagreement to the ignored interference effect which may be particularly important for the lowest order peaks, where the interfering contributions to the ionization amplitude may have comparable values resulting in strong interference modulations.

In conclusion, we have established a simple method for including Coulomb effects in the strong-field approximation and applied this method to describe direct ionization by elliptically polarized laser radiation. By comparison with new experimental data, the proposed model has been shown to work well except for ATI electrons of very low energy. The Coulomb force acting on the freed electron in the vicinity of the turning point, i.e., far away from the parent ion, is the key point for the mechanism of symmetry violation in our model. At the same time, that explains why one should not expect a pronounced effect of symmetry violation for the case of negative ions where the ionized electron interacts with the neutral residual atom via a short-range potential. The model can be generalized for larger systems such as molecules. We also note that the key processes underlying attosecond physics are subject to Coulomb effects and that the physical insight and the numerical tools provided here will help in understanding the corresponding effects better.

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