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Above-threshold ionization in the tunneling regime

A. Lohr, M. Kleber, and R. Kopold
Physik-Department T30, Technische Universität München, 85747 Garching, Germany

W. Becker*
Max-Born-Institut, Postfach 1107, 12474 Berlin, Germany

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A compact generalization of the Keldysh ionization amplitude is derived that includes rescattering. It is used for calculations of above-threshold ionization spectra with respect to energy for various emission angles for tunneling ionization of helium at high intensity, for the simple case of a zero-range potential as the binding potential. Most of the essential features of recent measurements are reproduced, that is, the onset, the extent, and the relative height of the plateau, which makes up the major part of the observed spectrum.
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Above-threshold ionization (ATI)—the phenomenon whereby an atom absorbs more photons than are actually necessary for ionization—has come a long way since its first observation nearly 20 years ago [1]; for a recent review, see [2]. As a consequence, the physical picture of the process of ionization has become much more colorful. In particular, the recent dramatic improvement of electron counting statistics owing to the development of high-repetition femtosecond lasers has revealed qualitative features [3–6] that look distinctive on a logarithmic scale, but would have escaped detection on the linear scale that necessity enforced in earlier days. The observations of the past few years have pointed to the significance of electrons returning to the ion for processes such as high-harmonic generation, double ionization, and, of course, above-threshold ionization itself [7]. In fact, on a logarithmic scale, the ATI electron spectrum at high energies consists of an extended plateau that owes its existence to rescattering.

The backbone of a compact theoretical description of ionization is the Keldysh theory [8], which satisfactorily accounts for a multitude of features of the electron spectra of ATI for comparatively low electron energies. In its common versions, however, it does not allow for rescattering. Hence, as it stands, there is no comparably compact expression that is capable of generating the entire ATI spectrum. In this Rapid Communication, we will derive such an expression and compare it to recent data taken at high laser intensity.

The strong-field approximation [9] within the Keldysh-Faisal-Reiss (KFR) framework has produced very good agreement with experimental data of strong-field ionization of helium [10,11]. However, these data did not extend to sufficiently high electron energies to display the rescattering-induced plateau, nor did the theory contain rescattering. The most recent measurements of Walker *et al.* [6] for helium at around 10^{15} W/cm^2 do reach up to electron energies exceeding $10U_p$ (the classical cutoff of the electron spectrum owing to rescattering [12]), and indeed they are, on the logarithmic scale, completely dominated by a very long plateau. Hence an extension of the standard KFR description is required. The plateau as well as the angular distributions of ATI at *moderate* intensities are well described by a three-step model that employs a zero-range potential for binding as well as rescattering [13]. A closely related description based on the first two steps of an iterative procedure for a Coulomb potential has been formulated in Ref. [14]; see also an approach along the lines of the Lewenstein model of high-harmonic generation [15]. A series of papers attempts to incorporate rescattering by using Coulomb-Volkov solutions instead of the ordinary Volkov solutions in the context of the standard Keldysh approach [16]. An approximation to multiple Coulomb scattering is proposed in Ref. [17]. Thus far, however, there is no calculation by any of these methods corresponding to the most recent high-intensity data of Walker *et al.* [6]. (We have presented preliminary results in Ref. [18].)

We proceed in the spirit of the usual Keldysh approximation, following a route that has already been applied to high-harmonic generation [19]. The matrix element for ionization from the ground state $|\psi_0(t)\rangle$ of an atom with binding potential V into a scattering state $|\psi_{\mathbf{p}}(t)\rangle$ with asymptotic momentum $\mathbf{p}$ is

$$M_{\mathbf{p}}=\lim_{t\to\infty,t'\to-\infty}\langle\psi_{\mathbf{p}}(t)|U(t,t')|\psi_0(t')\rangle, \tag{1}$$

where $U(t,t')$ is the time-evolution operator of the atom in the presence of the external laser field. It satisfies an integral equation which yields an expansion with respect to the interaction $H_I(t)$ with the external laser field ($\hbar=1$),

$$U(t,t')=U_0(t,t')-i\int_{t'}^{t}dt''U(t,t'')H_I(t'')U_0(t'',t'), \tag{2}$$

where $U_0(t,t')$ denotes the operator of free time evolution. We use this equation in the matrix element (1) and exploit the orthogonality of the ground state and the scattering state. We can then carry out the limit of $t'\to-\infty$ and arrive at

*Also at Center for Advanced Studies, University of New Mexico, Albuquerque, NM 87131.

$$M_{\mathbf{p}}=-i\lim_{t\to\infty}\int_{-\infty}^{t}dt'\langle\psi_{\mathbf{p}}(t)|U(t,t')H_I(t')|\psi_0(t')\rangle. \quad (3)$$

In addition to the integral equation (2) the time-evolution operator also obeys an integral equation with respect to the interaction with the binding potential V,

$$U(t,t')=U^{(V)}(t,t')-i\int_{t'}^{t}dt''U^{(V)}(t,t'')VU(t'',t'). \quad (4)$$

Here $U^{(V)}(t,t')$ represents the time-evolution operator of a free electron coupled through the interaction $H_I(t)$ to the external field laser field, viz., the Volkov time-evolution operator. Now, using the latter integral equation in the matrix element (3) we obtain two terms,

$$M_{\mathbf{p}}=-i\lim_{t\to\infty}\int_{-\infty}^{t}dt'\langle\psi_{\mathbf{p}}(t)|U^{(V)}(t,t')\{H_I(t')|\psi_0(t')\rangle$$
$$-i\int_{-\infty}^{t'}dt''VU(t',t'')H_I(t'')|\psi_0(t'')\rangle\}. \quad (5)$$

The first term yields the common Keldysh amplitude if we replace the scattering state $\langle\psi_{\mathbf{p}}|$ by a plane wave. It incorporates the atomic potential only in the initial state and is therefore not able to describe rescattering. In contrast, the second term allows for additional interactions with the atomic potential. In the representation (5), the matrix element is still exact.

In order to obtain a manageable expression we now replace in the second term the complete time-evolution operator U by the Volkov time-evolution operator $U^{(V)}$. Moreover, we rewrite Eq. (5) replacing in the second term $H_I(t'')=[\hat{\mathbf{p}}^2/2m+H_I(t'')]-[\hat{\mathbf{p}}^2/2m+V]+V$ and noticing that the two square brackets act like derivatives with respect to t'', respectively to the left and to the right. If now we integrate by parts, the contributions from the two square brackets cancel and the first term in Eq. (5) (viz., the standard Keldysh term) is canceled by a boundary term that occurs in this partial integration. Furthermore, we now replace the scattering state by a plane wave. We are then able to carry out the limit of $t\to\infty$ and are left with the compact result

$$M_{\mathbf{p}}=-\int_{-\infty}^{\infty}dt\int_{-\infty}^{t}dt'\langle\psi_{\mathbf{p}}^{(V)}(t)|VU^{(V)}(t,t')V|\psi_0(t')\rangle. \quad (6)$$

This expression takes into account both the "direct" electrons that depart from the atom without any further interaction with the binding potential as well as those electrons that rescatter. Following the same steps as above, the first term of Eq. (5) can be rewritten as

$$M_{\mathbf{p}}^{(0)}=-i\int_{-\infty}^{\infty}dt\langle\psi_{\mathbf{p}}^{(V)}(t)|V|\psi_0(t)\rangle, \quad (7)$$

which is an equivalent form [20] of the standard Keldysh amplitude. The state $\langle\psi_{\mathbf{p}}^{(V)}(t)|$, which appears both in Eq. (6) and Eq. (7) denotes the Volkov state, viz. the state of a free electron in a laser field with time-averaged momentum $\mathbf{p}$. Comparison of these two expressions shows that the former is an appealing generalization of the latter.

The time-evolution operator $U^{(V)}(t,t')$, which in Eq. (6) is sandwiched by the binding potential, allows for excursions of the electron away from and back to the ion. This becomes more transparent if the matrix element (6) is explicitly rewritten in position space,

$$M_{\mathbf{p}}=-\int_{-\infty}^{\infty}dt\int_{-\infty}^{t}dt'\int d^3\mathbf{r}d^3\mathbf{r}'\psi_{\mathbf{p}}^{(V)}(\mathbf{r},t)^*V(\mathbf{r})$$
$$\times U^{(V)}(\mathbf{r}t,\mathbf{r}'t')V(\mathbf{r}')\psi_0(\mathbf{r}',t'). \quad (8)$$

The propagator $U^{(V)}(\mathbf{r}t,\mathbf{r}'t')$ describes an electron propagating from the position $\mathbf{r}'$ at the time t' to the position $\mathbf{r}$ at the later time t where both $\mathbf{r}$ and $\mathbf{r}'$ are restricted to within the range of the atomic potential $V(\mathbf{r})$. In addition, the propagator also accomplishes some dressing of the initial and the final state.

The evaluation of the matrix element (8) becomes the simpler, the shorter the range of the atomic potential. In the limit of a zero-range potential, the two spatial integrations in Eq. (8) can be carried out trivially. Of the remaining two integrations over time, one yields the energy-conserving δ function. Hence, just one quadrature is left for numerical evaluation. The calculation is fairly straightforward and we are content with just presenting the final answer. The matrix element (8) is proportional to

$$M_{\mathbf{p}}\sim\sum_n\delta\left(\frac{p^2}{2m}+U_p+|E_0|-n\omega\right)\sum_{l=-\infty}^{\infty}J_{2l+n}\left(\frac{2p_x}{\omega}\sqrt{\frac{U_p}{m}}\right)$$
$$\times\int_0^{\infty}d\tau\left(\frac{im}{2\pi\tau}\right)^{3/2}\left(e^{-i[|E_0|\tau+l\delta(\tau)]}\right.$$
$$\times\exp\left\{-iU_p\tau\left[1-\left(\frac{\sin\frac{1}{2}\omega\tau}{\frac{1}{2}\omega\tau}\right)^2\right]\right\}$$
$$\left.\times J_l\left(y(\tau)\frac{U_p}{\omega}\right)-J_l\left(\frac{U_p}{2\omega}\right)\right). \quad (9)$$

Here $p_x=|\mathbf{p}|\cos\phi$ denotes the component of the electron's momentum parallel to the laser, U_p is its ponderomotive potential, $|E_0|$ stands for the binding energy, and the J_n are Bessel functions. The real quantities $y(\tau)$ and $\delta(\tau)$ are defined via

$$y(\tau)e^{-i\delta(\tau)}=\frac{1}{2}-i\left(\sin\omega\tau-\frac{4\sin^2\omega\tau/2}{\omega\tau}\right)e^{-i\omega\tau}. \quad (10)$$

The matrix element (9) includes the contribution of the direct electrons, viz., the standard Keldysh matrix element (7), whose explicit form is

$$M_{\mathbf{p}}^{(0)}\sim\frac{m}{2\pi}\sqrt{2m|E_0|}\sum_n\delta\left(\frac{p^2}{2m}+U_p+|E_0|-n\omega\right)$$
$$\times\sum_{l=-\infty}^{\infty}J_{2l+n}\left(\frac{2p_x}{\omega}\sqrt{\frac{U_p}{m}}\right)J_l\left(\frac{U_p}{2\omega}\right). \quad (11)$$

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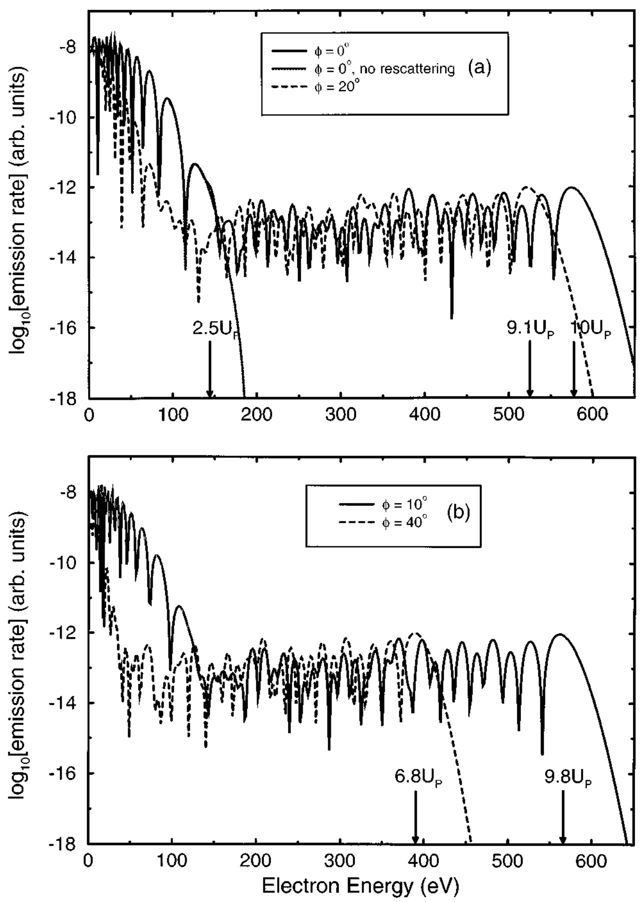

FIG. 1. Electron yields of ionization of helium by a linearly polarized laser with $\hbar\omega = 1.58$ eV at 10^{15} W/cm^2 at angles of (a) $\phi = 0°$ and 20° and (b) 10° and 40°, with respect to the polarization of the field. The arrows at $10U_p$, $9.8U_p$, $9.1U_p$, and $6.8U_p$ mark the classical end of the plateau for $\phi = 0°$, 10°, 20°, and 40°, respectively. The arrow at $2.5U_p$ in (a) points to the energy where the calculations based on Eqs. (6) and (7) start to differ from each other; that is, where rescattered electrons start to be more numerous than direct electrons. For $\phi = 0°$, the thin solid line in (a) gives the result of the standard Keldysh approximation (11).

The results (9)–(11) are very similar to an earlier version that was applicable in the multiphoton regime [13].

Figure 1 exhibits results of calculations based on Eq. (9). Electron spectra are shown for ionization of helium at 10^{15} W/cm^2 for $\hbar\omega = 1.58$ eV for emission at various angles ϕ with respect to the electric field of the laser. An extended plateau is the most prominent feature of all of the graphs. For each angle, the plateau has a very well defined cutoff. For emission along the direction of the field, the cutoff is at $10.007U_p$, as predicted by the completely classical model [12]. For the other angles considered, the cutoff energies as calculated from the same model are marked, respectively, by arrows. In each case, there is perfect agreement between this classical prediction and the fully quantum-mechanical calculation based on Eq. (9). At the intensity considered here, the drop of the plateau is much steeper than at the lower intensities, for which the plateau was originally discovered [4] and the first calculations were carried out [13]. For emission along the field, the result of the standard Keldysh amplitude (7) is also given, which describes only the direct electrons. It agrees precisely with the complete result up to about $2.5U_p$ (indicated in the figure), which is just below the onset of the plateau. From there on, the spectrum consists almost entirely of rescattered electrons. For emission off the direction of the field, the plateau starts for lower energies, as low as about U_p for $\phi = 40°$. For any angle, the plateau is very rugged. Its average elevation does not depend on the angle.

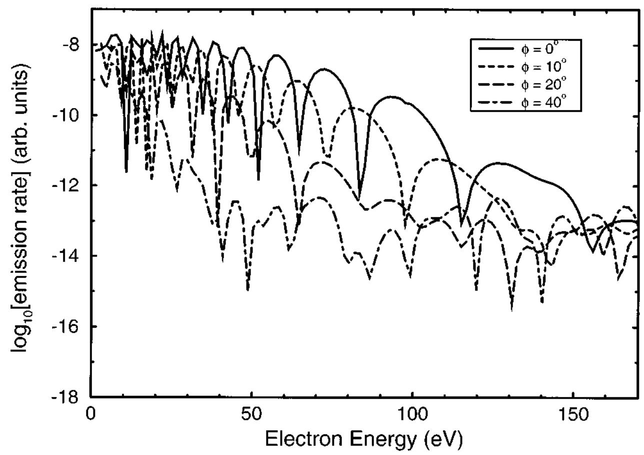

FIG. 2. Enlargement of the low-energy region of Fig. 1.

Figure 2 is an enlargement of the low-energy region of Fig. 1. It shows that for emission off axis the electron yield drops more and more quickly for increasing electron energy, as opposed to the conditions within the plateau. Another very conspicuous feature of the spectra is the narrow suppressions of the yield separated by fairly broad rounded tops, which are particularly well developed for $\phi = 0°$ and 10°. They are a manifestation of quantum-mechanical interference. For given energy and emission angle, in the tunneling regime electrons are released at precisely two times during one optical cycle. These two events interfere, and the interference alternates between constructive and destructive as a function of energy. In Fig. 2, we are concerned with the direct electrons, and their emission rate is proportional to the generalized Bessel function in Eq. (11). A saddle-point analysis of this expression produces exactly this sequence of constructive and destructive interferences and supports the interpretation in terms of tunneling interferences [21]. Our results are based on the assumption of constant intensity, a condition that in experiments in the tunneling regime one will hardly be able to meet. Hence, these interferences may never show up in an actual experiment and, indeed, they are not visible in the available data of Ref. [6]. There is, however, a closely related situation where they have been seen already, again in close agreement with theory. This is in the ellipticity dependence of the ATI spectra at fixed energy [21]. Ellipticity provides a tunable parameter that can be kept constant throughout the pulse.

For any angle, the plateau is made up of a sequence of sharp suppressions and rounded tops much like the spectrum of the low-energy direct electrons. The origin is likely again to be interference of tunneling trajectories as suggested for the plateau in high-harmonic generation [22], calculations of which look very similar. As opposed to the direct electrons, the interferences within the plateau are not as easily pinned down quantitatively, owing to the more complicated mechanism of rescattering. Just before the end of the plateau, the final rounded top is the largest of all. This region is the most classical part of the plateau.

Comparing our calculations to the experimental data of Walker *et al.* [6] we observe good qualitative agreement with respect to the existence and the extent of the plateau, its relative height as compared to emission at low energy, and the angular dependence of the emission of the direct electrons. There are two apparent discrepancies. First, the calculated plateau is horizontal as opposed to the measured one, which slopes downward. Averaging the calculated spectrum over the intensity distribution in the laser pulse would introduce such a downward slope. Second, the height of the measured plateau drops with increasing angle of emission, while the height of the calculated plateau is largely independent of this angle. Again, the averaging would remove part of this discrepancy, but partly it is likely to be due to the properties of the zero-range potential. This potential scatters isotropically, which gives more emphasis to large scattering angles than a more realistic potential would.

We have, in this paper, derived a generalization of the standard ionization amplitude of the Keldysh approach that incorporates a single return of the electron to its parent ion and, therefore, allows for rescattering. It holds for an arbitrary binding potential, but, as the ordinary Keldysh amplitude, will work the better the higher the intensity of the laser field and the shorter the range of the binding potential. We have explicitly calculated electron spectra for ionization of helium at 10^{15} W/cm^2 and obtained very good qualitative agreement with the recent data of Walker *et al.* [6]. Our results lend additional support to the conclusion that for many phenomena in high-intensity laser-atom physics the detailed shape of the atomic potential is not essential.

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