Collective Multielectron Tunneling Ionization in Strong Fields

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We investigate the quantum mechanical process of two-electron tunneling in strong external electric fields. Numerical solution of a two-electron *s*-wave model reveals the existence of collective tunneling ionization in a mode where both electrons stay at equal distance from the nucleus. Otherwise the lagging electron is immediately recaptured. The corresponding double ionization rate fails to explain nonsequential multiple ionization in strong-field laser experiments. However, an empirically modified version of the analytical one-electron tunneling rate of Ammosov, Delone, and Krainov agrees with the experiments to a surprising accuracy. The reason for this agreement is presently unknown.

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The phenomenon of one particle tunneling through a potential barrier is one of the most important ionization mechanisms in strong external fields [1–3] and has been extensively investigated. In contrast, simultaneous tunneling of more than one particle has, to our knowledge, never really been considered in atomic physics. This is the more surprising since atomic collective many-particle effects and correlations are generally under intensive investigation. For instance, electron-electron correlations are usually considered responsible for the phenomenon of "nonsequential" or "simultaneous" multiple ionization in strong laser fields. A number of simple models [4–10] and sophisticated numerical calculations [11–13] have been applied, but the question of the exact physical mechanisms behind nonsequential ionization is still not entirely settled.

In this Letter, we address the question whether the process of simultaneous tunneling of electrons in external fields exists and may help to explain the experimental double ionization results. This could be expected since one-electron tunneling is commonly accepted as the reference process in single ionization at sufficiently strong fields. Our investigation was stimulated by the observation that an empirical, yet unexplained modification of the one-electron ADK (Ammosov-Delone-Krainov) [3] tunneling formula reproduces most of the experimental data on nonsequential ionization. For a rather comprehensive answer to this question we take the following steps: First, we investigate the fundamental quantum mechanical problem of two-electron tunneling in static electric fields. Both from analytical model calculations and from the numerical solution of a (1D $\times$ 1D) Schrödinger equation we find that (i) collective multielectron tunneling ionization (CTI) does exist in static fields, and (ii) proceeds through a delicate dynamical balance in a truly collective mode. Second, we find that the calculated static two-electron tunneling rate is far too small to explain the experimental nonsequential laser ionization for helium. Only at shorter pulse durations (below the fs range) the CTI process may substantially exceed the sequential double ionization. Finally, we analyze the empirical formula for the nonsequential ionization rate in strong laser fields in the light of the quantum mechanical CTI results. This raises both new insights and new questions, but fails to link the formula with a true tunneling process, despite close similarities in the mathematical structure. Thus, the origin of the empirical formula remains a puzzle after all. However, we emphasize the empirical formula as a very useful pragmatic tool to quantitatively estimate multielectron ionization in strong laser fields, and as food for further thought.

We first turn to the fundamental quantum mechanical problem of simultaneous two-electron tunneling in the quasistatic approximation. We start by recalling the single-electron ionization rate in the tunneling regime by an ac field with amplitude F, as given by the ADK formula [3]:

$$W_{\mathrm{ADK}} = A\left(\frac{4\sqrt{2m_e E_i^3}}{e\hbar F}\right)^B \exp\left(-\frac{4\sqrt{2m_e E_i^3}}{3e\hbar F}\right), \qquad (1)$$

where m_e, e, and E_i denote the electron's mass, the absolute value of its charge, and its binding energy in the state $i \equiv (n^*, \ell, m)$, respectively. The quantities A and B are known in analytical form and depend only on E_i and (n^*, ℓ, m). The dependence of the ionization rate on $E_i^{3/2}/F$ is dominated by the exponential factor.

Naively, one may envision CTI of N electrons as tunneling of one hypothetical quasielectron of mass Nm_e, charge Ne, and total binding energy $E_{\mathrm{tot}} = \Sigma_N E_j$. Assuming that all of the kinetic energy is in the center-of-mass motion one obtains a CTI rate proportional to $\exp\{-4(\sqrt{2m_e E_{\mathrm{tot}}^3/N})/3eF\}$. Zon [14] arrives at the same result via a more elaborate calculation which also yields the correct form of the prefactor. Defining $E_{\mathrm{eff}} = E_{\mathrm{tot}}/N$, the exponential can be rewritten as $\exp\{-4N(\sqrt{2m_e E_{\mathrm{eff}}^3})/3eF\}$. Thus, considering all prefactors, this two-electron tunneling rate turns out to differ from the one-electron ADK formula essentially in two points: an additional factor N in the exponential, and the consistent substitution $E_i \rightarrow E_{\mathrm{eff}}$.

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This analytical result was obtained under the assumption that the electrons behave like a quasiparticle of N times the mass and charge throughout the process, which is by no means *ab initio* evident. Therefore we have performed numerical calculations in order to reveal the underlying dynamics of the process. It is sufficient to restrict oneself to static fields and one dimension (1D) per electron, for which we choose a spherical s-wave model. The three-dimensional degrees of freedom of real electrons are mimicked by this model since the interelectronic repulsion is mitigated. It has been used widely in the analysis of electron scattering and is a simple example of a more realistic, smoothed Hartree model. The Hamiltonian in the two radial variables $x > 0$ and $y > 0$ is

$$H(x,y) = T + V(x) + V(y) + V_{12}(x,y) - eF(x+y), \tag{2}$$

where $T = T_x + T_y$ is the total kinetic energy, $T_x = p_x^2/2m_e$, and the nuclear potential is given by $V(x) = -eZ/x$ with $Z = 2$. The interelectronic repulsion leads to a screening of the nuclear charge for the outer electron, thus $V_{12}(x,y) = +1/\max(x,y)$. The external static electric field F points radially outward. This does not appear to be realistic; however, we take this model as an extreme case in which the interelectronic interaction term can never increase the force pushing the "inner" electron towards the nucleus, in contrast to collinear 1D models in which $V_{12} = V(|x-y|)$. Numerical results for the ionization rates are obtained from the stationary complex-energy eigenvalues and eigenvectors represented on a position space (x,y) grid including absorbing boundary conditions by means of a complex absorbing potential [15]. From the wave function we extract the local probability density flux $\mathbf{j}(x,y) = \mathrm{Im}[\Psi^*(x,y)\nabla\Psi(x,y)]$, visualized in Fig. 1.

The flux diagram reveals the dynamics of the two-electron tunneling ionization in static fields within the limits of our model. Most of the ionization proceeds parallel to the axes at small x or y, which corresponds to single ionization. We note that we always expect a nonzero, although very small component of these flux arrows into the orthogonal direction, corresponding to tunneling of the remaining tightly bound electron in the sequential ionization process at large x/small y (or large y/small x). Flux vectors with considerable nonzero components in both coordinates correspond to nonsequential (or collective) tunneling of both electrons. We observe two key elements of the collective tunneling dynamics: first, there is considerable flux emerging near the origin into the direction of the main diagonal ($x = y$), which, however, weakens quickly as the electron pair moves away from the nucleus. Second, at larger distances from the nucleus the flux exhibits a "bistable" behavior: it either follows closely the main diagonal, indicating a highly correlated motion of the electrons with $x \approx y$, or it turns parallel to one of the axes, indicating capturing of one of the

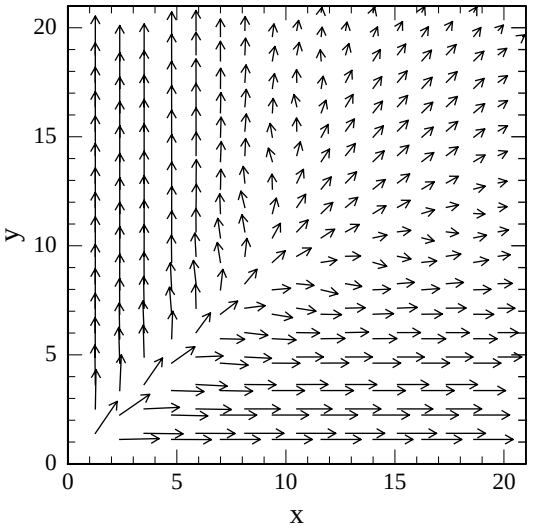

FIG. 1. Probability flux density of static-field ionization for the Hamiltonian (2) at a field strength $F = 0.18$ a.u. The arrows give the direction of the flux $\mathbf{j}$ and their length is proportional to the logarithm of $|\mathbf{j}|$, spanning 12 orders of magnitude. The picture is not symmetric under interchange x–y, although the wave function is, since the positions of the vectors have not been chosen symmetrically.

electrons in a state with constant radial coordinate. Hence, we conclude that simultaneous two-electron tunneling proceeds only through a highly correlated, although unstable collective mode where the electrons remain close to each other, thus partially screening the nuclear charge from each other. Otherwise, the lagging electron is immediately recaptured into a quasibound state, from which it tunnels with a much reduced rate in a sequential manner. We have performed similar calculations for the widely used collinear model with $V(x,y) = 1/\sqrt{(x-y)^2 + a}$ and $-\infty < x, y < +\infty$. The flux picture looks very similar to the one in Fig. 1. Thus, the double ionization flux again can only leave the nucleus near the diagonal, $x = y$. At large x and y, however, the electronic repulsion pushes the electronic density away from the diagonal. This does not lead to the recapture of the inner electron if both x and y are large enough (i.e., the electrons have left the double ionization tunnel barrier).

The total ionization rate is given by the total flux leaving the region at large values of (x,y). The border between the two asymptotic flux directions gives an operational distinction between the single and the double ionization rate, the double ionization rate being many orders of magnitude smaller than the rate for single ionization. The collective double ionization flux turns out to be in good agreement with the analytical formula by Zon [14], but much too low to explain the nonsequential ionization yield in strong laser fields. In fact, the calculated doubly charged-ion yield through CTI at the commonly used pulse lengths is only slightly larger than the calculated yield through a sequential tunneling ionization process. Thus, the relevance of CTI appears to be considerably less than other mechanisms, notably the so-called "rescattering" model,

even though the latter also appears to lie well below the experimental results [10]. Hence, we conclude that CTI, although it clearly exists, does not explain the experimental data on nonsequential ionization in strong laser fields.

There are, however, several situations where CTI is the dominant mechanism of double ionization in a static field, at least when saturation effects can be completely neglected. First, recalling that there is no single *rate* for sequential ionization, we note that any comparison between sequential and nonsequential ionization depends on the observation time window. In particular, for sufficiently short times CTI will always be dominant, no matter how small it is. Second, this time domain can be substantially prolonged if one ionization potential is much smaller than the other, such as is the case in H^-. Then, owing to the nonlinear dependence of the exponent in the ADK formula on the binding energy, the CTI rate may even exceed the ionization rate of the second electron, the bottleneck in the sequential process, at any relevant field strength. For normal ground state atoms, such as the noble gases, CTI exceeds the second electron's ionization rate only in the limit of very high field strengths, as first pointed out by Zon [14].

With this theoretical background we now return to our empirical formula. It is a fact that a simple empirical modification of the one-electron ADK tunneling rate (1) yields surprisingly good agreement with experimental N-electron ionization in strong laser fields. Technically, the modification consists of the mere replacement of the one-electron binding energy E_i in the ADK rate (1) by the effective one-electron binding energy E_{eff}. The effective quantum number n^* is given by $E_{\mathrm{eff}} = Z^{*2}/2n^{*2}$, where $Z^* = Z - \sigma$ is the effective nuclear charge. We use a shielding factor $\sigma = 0.3$ [16] in all of our calculations.

We emphasize that a deviation of only a few percent from the correct value of E_{eff} completely destroys the agreement. In the light of collective tunneling, we have seen that the dependence on E_{eff} is a characteristic feature which follows directly from the N-electron tunneling dynamics: Since the electrons must stay closely together in order to escape, the amount of work spent on each electron in an equivalent zero-field condition, i.e., the "zero-field binding energy," is exactly E_{tot}/N. On the other hand, starting from the correct CTI rate we are forced to drop a factor N (where N is the number of electrons) in the exponential in order to arrive at the empirical formula. Neither the ADK rate modification nor the CTI rate modification has yet been justified theoretically, but each one appears severe enough to rule out CTI—or even tunneling at all—as the underlying physical mechanism, despite the apparent close relation in the mathematical structure.

For illustration, in Fig. 2 we compare the results of the empirical formula to experimental ion yields. The solid curves have been calculated from rate equations in the usual manner [9]. We note that for any given atom there exists only one free parameter in order to adjust one (and only one) charge state yield for one single intensity to the experiment.

Figures 2(a) and 2(b) show the double ionization of helium and neon ($E_{\mathrm{eff}} = 39.4$ and 31.3 eV, respectively) [7,9]. In both cases the characteristic double ionization structure ("knee") lies several orders of magnitude above the sequential result (dotted line), and is reasonably well reproduced by the empirical formula (solid line). In neon a small deviation is obvious in the vicinity of the knee, but is not more than a factor of 3. In helium, the formula deviates progressively towards lower intensities, while in neon it nicely follows the experimental data over 7 orders of magnitude.

We have also performed first ionization yield measurements on metal vapors [17] to test the empirical formula beyond closed-shell rare gases. Specifically, we investigated the double ionization of In^+ (a heliumlike system with similar valence electron binding energy E_i, but different E_{eff}), for which we found the predictions to be very well confirmed. Multiple electron ionization ($N > 2$) in the heavy rare gases Ar, Kr, and Xe [9] is shown in Figs. 2(c)–2(e): A good, in some cases perfect, agreement for the doubly and triply charged ion yields is achieved,

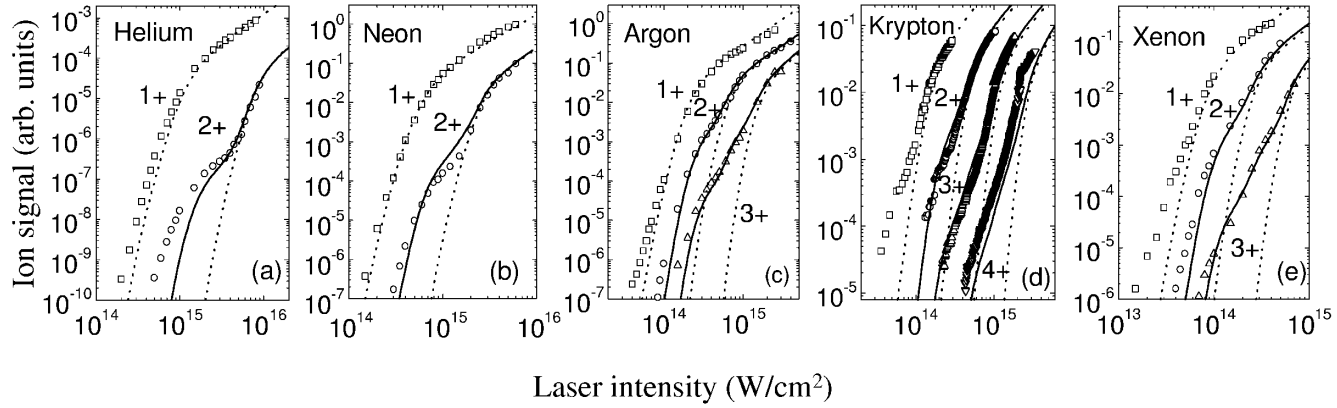

FIG. 2. Multiple ion yield in a laser pulse. (a) Helium, experimental data from [7]; (b) neon, (c) argon, and (e) xenon, experimental data from [9]; (d) krypton, experimental data from [17]. Markers: experimental data; full curve: empirical formula, using rate equations; dotted curve: ADK formula using purely sequential ionization.

extending in Kr even to Kr^{4+} data, obtained at our laboratory [17]. Here the application of the empirical formula to multiple ionization reveals additional information on the ionization mechanism, namely, the pathways in the multiple ionization cascades. For instance, in the case of the Kr^{4+} ion yield, the simultaneous double ionization of Kr^{2+} is dominant only in the vicinity of the saturation intensity of Kr^{3+}, at about 10^{15} W/cm^2, whereas the other intensity regions are dominated by simultaneous triple ionization of Kr^{+}.

Interestingly, we have successfully reproduced experimental ionization yields for different laser frequencies, although the empirical formula, since it is based on the ADK rate (1), is inherently independent of the laser wavelength. Experimental results on multiple ionization of argon at 1053 nm [9] and helium at 248 nm [18], which differ from the results at 800 nm, are again satisfactorily described. Obviously, the different yield curves are simply due to the longer laser pulse duration of about 500 fs in both experiments, which substantially alters the ion yields (as confirmed by the rate equations) even though the ionization rates themselves are unchanged. Furthermore, we confirm the absence of a knee structure in the ion yield data for ionization of Xe and Kr in a strong CO_2 laser field at 10 μm [19]. Our calculation predicts that the knee should appear just below the laser intensity range studied in Ref. [19].

There is a systematic underestimate of the experimental rates at lower laser intensities, which is expected from the exponent in the empirical formula, as opposed to the experimentally observed weak-field behavior of nonsequential ionization [7,9]; incidentally, the deviation is most pronounced in the case of helium double ionization. Similar asymptotic deviations are observed in true one-electron tunneling processes. Finally, we point out that we have not considered the possibility of extending the empirical formula to cover the case of elliptical polarization of the laser field.

In conclusion, there are two main messages of the present Letter. First, we have investigated the dynamics of the two-electron tunneling process in external electric fields, a phenomenon which has received little or no attention in the past, although being of fundamental nature. We find that two-electron tunneling exists, and is dominated by a highly correlated mode where the two electrons remain at equal distances from the nucleus, otherwise the lagging electron is immediately recaptured. The total ionization rate of this collective tunneling mode is small; in particular, it is far too small to account for the observed nonsequential double ionization in external laser fields. Second, we found a simple empirical formula for N-electron ionization in strong laser fields, which reproduces most of the existing experimental data for linear laser polarization to a surprising accuracy and without free parameters over several orders of magnitude below saturation intensity. The origin of this agreement and, in particular, the theoretical background for this formula are unknown at this time. However, such a simple analytical expression is interesting in its own right as it greatly facilitates, e.g., large-scale plasma computations, if the limits of applicability are reliably known.

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