δ ionization: Stratified symmetrical electron emission and resonantly structured ionization continuum

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We have discovered theoretically that ionization of a one-dimensional quantum well by an ultrashort subcycle (δ-like) electromagnetic pulse results in photoelectrons propagating both along the field and in the opposite direction in well-separated shells whose velocities reflect quasienergies in the ionization continuum. For both a quantum well and a three-dimensional hydrogenlike atom, a unipolar, i.e., strongly asymmetrical, δ ionization produces an approximately symmetrical, stratified photoelectron cloud.

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The theory of photoionization, one of the most fundamental and rich phenomena in atomic physics, was substantially revised for high laser intensities. In particular, Einstein’s formula for the photoelectron kinetic energy, \( E_k = \hbar \omega - E_{\text{ion}} \), while valid for relatively weak monochromatic fields, breaks down in laser fields strong enough to cause ionization even if the photon energy \( \hbar \omega \) is much smaller than the ionization energy \( E_{\text{ion}} \). Almost all strong-field effects discovered so far (above-threshold ionization, barrier-suppression ionization, and atomic stabilization) to name a few [1]) are, however, caused by quasimonochromatic oscillating electric fields of lasers. Recently [2], we theoretically demonstrated the feasibility of intense ultrashort (near-femtosecond or subfemtosecond) subcycle pulses which, we believe, call for yet another important extension of the theory of interaction of electromagnetic radiation with matter. Indeed, in contrast to the most commonly studied optical fields, no particular frequency could be ascribed to a subcycle pulse; the notion of a pulse envelope becomes equally useless. Together with the feasible ultrashort time duration, this renders irrelevant such common tools of the theory as the rotation-wave and slow-varying-envelope approximations. Moreover, for a subcycle pulse shorter than typical times of a quantum system (such as the ‘‘ionization time’’ \( t_{\text{ion}} = \hbar / E_{\text{ion}} \), where \( E_{\text{ion}} \) is the ionization potential of the system), the pulse amplitude and duration are no longer meaningful if taken separately. Instead, the pulse’s effect on the system is solely determined by the pulse area, \( Q = \int dt \varepsilon (t) \) \( \varepsilon (t) \) being the pulse’s electric field; \( Q \), in turn, is proportional to the average momentum imparted by the pulse (by contrast, \( Q \sim 0 \) for an oscillating pulse). Indeed, such pulse may be modeled by the δ function of time. Then the wave functions just before (\( \Psi_0 \)) and after (\( \Psi \)) the pulse are related simply as \( \Psi = \exp (-i Q / \hbar) \sum_{\tau} \Psi_0 \), where the sum is taken over all the charged particles in the system [3]. It is easy to see now that the average momentum of the system changes by \( \Delta p = e Q \), while the change in the average total energy looks like a classical relation between the changes in the momentum and kinetic energy: for a one-particle system, for simplicity, \( \Delta E = \int dx \Psi^* H_0 \Psi - \int dx \Psi_0^* H_0 \Psi_0 = (\Delta p)^2 / 2m + p_0 \Delta p / m \), where \( p_0 \) is the average momentum of the system in the initial state, and \( H_0 \) is the system’s Hamiltonian without a subcycle pulse (field-free Hamiltonian). (This classical-like behavior is apparently due to the fact that the system does not have time to move during an ultrashort pulse.) One important consequence of this role of the pulse area is that the definition of the high-field regime, based on comparing the pulse field with the “atomic” field \( \sim \sqrt{E_{\text{ion}}} \) is not applicable to δ-like pulses.

Besides their apparent theoretical importance, ultrashort subcycle pulses of high intensity, being capable of strongly exciting or ionizing many quantum systems almost instantaneously on the systems’ time scale, may become an important new probing tool and a source of new effects in nonlinear optics and atomic and molecular physics. So far, however, δ ionization (and δ excitation) remains largely unexplored. One important exception is ionization of Rydberg atoms by almost unipolar, “half-cycle” pulses of the duration (~0.5 ps) comparable to Rydberg orbital times, which differs substantially from the ionization by lasers or by longer field pulses [4]. In this paper, we predict a qualitatively new effect, to our knowledge. We show, theoretically and by computer simulation of a one-dimensional (1D) quantum well (QW) and a 3D hydrogenlike atom, that the photoelectron cloud produced by δ ionization displays a strongly stratified spatiotemporal structure [5]: electrons are emitted in well-separated bunches both in the direction of the pulse field and in the opposite direction. For a relatively small pulse area, the photoelectron cloud is approximately symmetrical in regard to the pulse-field direction; this symmetry vanishes for higher amplitudes.

An important distinction of our approach is our focus on the photoelectron wave function in the coordinate representation. This focus is productive because of the unique, “trans-spectral” coherence of the δ ionization, as opposed to the ionization by a white-noise signal of a long timewidth with an equally broad spectrum. It is this coherence that maps the energy-level structure of a quantum system into the ordered spatiotemporal structure of the photoelectron cloud, rather than into a random superposition of electron bunches that would result from a white-noise ionization. While the ionization of Rydberg atoms by a δ-like electrical field “kick” has been considered before [6], in particular when a

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spatially uniform $\delta$ function of time was used to approximate the field of a fast incident electron [7], no attempts have been made to predict the spatiotemporal behavior of photoelectrons. We are unaware of any research on $\delta$ ionization of a QW.

The Schrödinger equation for an electron interacting with the pulse electric field $\varepsilon(t)$ is

$$i\hbar \partial \Psi(i,r)/\partial t = [H_0(r) - e r \cdot \varepsilon(t)] \Psi(i,r),$$  

(1)

$H_0$ being the field-free Hamiltonian. We assume here that the pulse field is uniform across the quantum system (the dipole approximation). One can show that this assumption is justified for $\delta$-like pulses if $E_{\text{ion}} \ll mc^2$, which coincides with the limits of nonrelativistic quantum mechanics and is apparently the case for any atom, and even more so for QW’s. Modeling $\varepsilon(t)$ by the Dirac $\delta$ function of time, $\varepsilon(t) \rightarrow \varepsilon_0 \delta(t/\tau_p)$, $e_{\varepsilon} \tau_p = \int_{-\infty}^{\infty} \varepsilon(t) dt = eQ$, one obtains a simple relation between the wave function before, $\psi_0$, and immediately after the pulse (at $t = 0^+$) as (see, e.g., Ref. [6])

$$\Psi(0^+) = \psi_0(r) \exp(i p_\varepsilon \cdot r/\hbar),$$

$$p_\varepsilon = e_{\varepsilon} \tau_p = \int_{-\infty}^{\infty} \varepsilon(t) dt = eQ,$$

(2)

where $p_\varepsilon$ is the classical (equal to the quantum average) momentum transfer due to the $\delta$ kick. Equation (2) is obtained by neglecting $H_0$ in Eq. (1) in the presence of the pulse field (the impulse approximation [3,6]).

The evolution of the system at $t > 0$ is governed by the field-free Schrödinger equation, with $\Psi(0^+)$ as the initial function. We express solutions to this equation through the full orthonormal set of bound $[\psi_j(r), \text{with } E_j \text{ as the eigenvalue}]$ and continuum $[\psi_c(r)$, for a given energy $E]$, eigenfunctions of $H_0$ as

$$\Psi(t,r) = \sum b_j \psi_j(r) e^{-iE_jt/\hbar} + \psi_{\text{ion}},$$

(3)

$$b_j = \int d^3r \psi_j^*(r) e^{ip_\varepsilon \cdot r/\hbar} \psi_0(r),$$

where $\Sigma$ stands for the summation over the bound states. Note that for weak $\delta$ kicks, $b_j \approx i p_\varepsilon d_{1j}/\hbar = i \phi_j^{(R)}$, where $d_{1j} = \int \psi_0^*(r) \psi_j^*(r) d^3r$ is the transition dipole moment between the initial state and the $j$th eigenstates, and $\phi_j^{(R)}$ is the Rabi phase. Of primary interest to us is the “ionized part” of the total wave function, $\Psi_{\text{ion}} = \int dE \ b(E) \psi_c(r) e^{-iEt/\hbar}$, that comprises the eigenfunctions of the continuum and, therefore, describes the motion of photoelectrons. We investigate in more detail two fundamental quantum systems: a 1D quantum well (QW) and a 3D hydrogenlike atom. Much of the symmetry of the $\delta$ ionization, however, is not related to particular potentials. It is easy to show, e.g., that for any real and inversion-symmetrical $H_0$, immediately after a highly asymmetrical (unidirectional) $\delta$ kick, the spatial distribution of photoelectrons is exactly symmetrical with regard to the field direction. Indeed, bound eigenfunctions $\psi_j(r)$ of such $H_0$ can always be chosen to be real and of a definite parity, with the ground-state wave function being even; assume that $\psi_0$ is even. Furthermore, the amplitudes $b_j$ are real (imaginary) for even (odd) $\psi_j$, so that at $t = 0^+$, Eq. (3) can be written as

$$\Psi_{\text{ion}}(0^+, r) = M(r) + iN(r),$$

$$M(r) = \left[ \cos(p_\varepsilon \cdot r/\hbar) - b_0 \right] \psi_0 - \sum b_{j\text{even}} \psi_{j\text{even}},$$

$$N(r) = \sin(p_\varepsilon \cdot r/\hbar) - \sum b_{j\text{odd}} \psi_{j\text{odd}},$$

where both $b_{j\text{even}}$ and $b_{j\text{odd}}$ are real, as are $M$ and $N$. Since $M(-r) = M(r)$ and $N(-r) = -N(r)$, $|\Psi(0^+, r)|^2 = M^2 + N^2$ is an even function of $r$ and, therefore, represents an inversely symmetrical photoelectron distribution. Intuitively, this symmetry might be expected, as the initially symmetrical electron distribution is instantaneously ionized by a uniform field.

Furthermore, the photoelectron propagation is approximately symmetrical with regard to the field direction. The symmetry is purely quantum. Indeed, a classical particle hit by the unidirectional field of a sufficiently strong $\delta$ kick would move in the direction of the force. Quantum $\delta$ ionization, on the other hand, is similar to hitting water in a shallow bucket: a kick would spill water all over the rim, unless the kick is very strong, in which case almost all the water will move along the kick. (Similarly, the scattering of a particle off a power center is isotropic for small incident momenta [3].) This symmetry holds for any inversion-symmetrical Hamiltonian, but is easier to see in one dimension. The continuous spectrum of such a 1D Hamiltonian is doubly degenerate. We may choose the eigenfunctions $\psi_{\pm}(x)$ for a given energy $E$ to mirror each other: $\psi_-(x) = \psi_+(x)$. Then

$$\Psi_{\text{ion}}(t,x) = \int_0^\infty dE \left[ b_+ \psi_+ + b_- \psi_- \right] e^{-iEt/\hbar},$$

(5)

$$b_+ = \int dx \ \Psi(0^+) \psi_+^2.$$  

For weak kicks, i.e., when $ap_\delta/\hbar \ll 1$ (a being a characteristic size of the system), we have

$$b_+(E) \approx -b_-(E) \approx i(p_\varepsilon/\hbar)d_{j,E},$$

$$|b_+(E)|^2 \approx |b_-(E)|^2,$$

(6)

so that the photoelectron spectra are almost identical to the spectrum of the dipole matrix elements of the field-free Hamiltonian, while photoelectrons are almost equally likely to propagate in both directions. To avoid misunderstandings, it is important to keep in mind that even a weak kick, i.e., a $\delta$ pulse of a small area, may create photoelectrons with very high energy and momentum, although rarely. Indeed, the energy spectrum of photoelectrons, according to Eq. (6), is close to the dipole momentum spectrum of the QW, which spreads, strictly speaking, to infinitely high energy (note also that the energy spectrum of a $\delta$ function is flat). For example, under the conditions of Fig. 2 below, the kinetic energy of about 4.6% of emitted electrons is larger than the depth of the quantum well.
This approximate “forward-backward” spectral symmetry holds for strong kicks as well, although becomes less exact. Combined with the above choice of the eigenfunctions, it leads to the spatial symmetry of the expanding photoelectron cloud. For detailed consideration, we now turn to two particular quantum systems.

(i) One-dimensional quantum well (QW). We consider a quantum well in the idealization whereby no solid-state effects are taken into account; we believe, however, that the qualitative picture we predict here will largely hold for a real semiconductor structures. A 1D quantum well in the x axis is defined by the potential $U = -U_0 = \text{const} < 0$ at $|x| < a$, and $=0$ elsewhere. We choose $\psi_+ \uparrow$ as a plane wave that propagates from left to right, with both incident and reflected waves on the left of the well, and only one (transmitted) right to it [8]. Hence

$$\psi_+(x < a) = e^{ik_x x} + A e^{-ik_x x},$$
$$\psi_+ (|x| \leq a) = B e^{ik_x x} + B_+ e^{-ik_x x},$$
$$\psi_+ (x > a) = C e^{ik_x x},$$

$$k_x = \sqrt{2mE/h}, \quad k_u = \sqrt{2m(E + U_0)/h}.$$

Similarly, $\psi_-(x) = \psi_+(-x)$ propagates from right to left. The constants $A, B, C$, and $C$ are found from the continuity of $\psi_\pm$ and $d\psi_\pm/dx$ at $x = \pm a$ as

$$A = k_0^2 (\gamma_u^2 - \gamma_U) \gamma_U^2 D^{-1},$$
$$B_\pm = 2k_E (k_U \pm k_E) \gamma_U \gamma_U^2 D^{-1},$$
$$C = 4k_E k_U \gamma_U^2 D^{-1},$$

$$D = (k_0 + k_E)^2 \gamma_U^2 - (k_0 - k_E)^2 \gamma_U^{-2} k_0 = \sqrt{2mU_0}/h,$$

$$\gamma_{U(U)} = e^{-iak_{E(U)}},$$

and $|A|^2 + |C|^2 = 1$. The QW is fully “transparent,” $|C|^2 = 1$, if

$$2ak_U = n\pi, \quad n \text{ integer};$$

in this case, the amplitude of the wave function inside the well is maximal, $|\psi_\pm|^2 = |1 + k_0^2 \sin^2(2ak_U/4k_E^2)k_0^2|^{-1}$. The respective resonant energies

$$E_n = (n/S)^2 - U_0, \quad n \geq S = 2ak_0/\pi$$

coincide with the positive eigenenergies of an infinitely high well of the width $2a$, whose walls begin at the energy $-U_0$. ($S$ characterizes the binding strength of the QW, as $[S] + 1$ is the number of the bound states in the well; $[S]$ is the integer part of $S$.) Maxima of the QW transparency, in effect “continuum shadows” of the QW, are at the core of the bunchlike ionization of the QW. This effect, however, is not simply a consequence of the transparency resonances [Eq. (10)]. Indeed, while even and odd $n$ in Eq. (10) both correspond to the zero reflectivity of a QW, they play drastically different role for $\delta$ ionization, due to the following easily verifiable selection rule: $|d_{1,E}\uparrow|^2$ zeros out for even $n$ and reaches local maxima at approximately $E_{n \text{ odd}}$ (again, in full analogy with the dipole selection rules in the infinite QW). (For a QW with $[S] + 1$ even, an extra maximum appears in the dipole spectrum just above the bottom of the ionization continuum, below the lowest resonance allowed by Eq. (10), as a result of the fact that the continuous function $|d_{1,E}\uparrow|^2$ is equal to zero at both $E = 0$ and at the lowest “regular” resonance.)

Weak $\delta$ kicks simply map this structure into the photoelectron energy spectrum. As the kick area grows, the spectral maxima and minima shift from Eq. (10) to higher (lower) energies for forward- (backward-) moving electrons—the effect that could be viewed as a $\delta$-ionization analog of the Stark shift. Figure 1 shows that the momenta $k_\uparrow(\pm)^{\min}$ for which $|b_\downarrow(E)|^2 = \min$, depend almost linearly on the (normalized) momentum transfer $q = p_s a/h$: $[S] = 7$ is assumed.

$$ak_\uparrow^{\min}(\pm) \approx \pm q + n\pi/2,$$

$$n = n_{\min}, n_{\min} + 2, \ldots \quad (11)$$

where $n_{\min}$ is the smallest even $n$ allowed by Eq. (10). The maxima (except for the above-mentioned near-bottom one) follow that pattern as well, with more pronounced but diminishing oscillations. Physically, this means that the velocities of the photoelectron bunches are determined, for a given QW, by the pulse area, so that Eq. (11) might be used to measure areas of ultrashort pulses. (The oscillations in Fig. 1 are reminiscent of the behavior of quasienergies in a superposed two-level system [9].) While Fig. 1 (and Fig. 2 below) has been obtained for a QW with $[S] = 7$, we have observed qualitatively similar pattern for $1 \leq [S] \leq 30$. Equation (11) becomes almost obvious for kicks so strong that most of $\Psi_{\text{ion}}$ comes from the high-energy eigenfunctions which can be approximated by plane waves; then $b_\downarrow(k_U) \sim \psi_0(\pm k_U - q)$, where $\psi_0(k)$ is the initial wave function in the momentum representation. As an important consequence, the photoelectron spectrum after a strong $\delta$ ionization provides a “snapshot” of $\psi_0$.

The $\delta$ pulse translates the structured ionization continuum into a spatial stratification of the photoelectron cloud. This phenomenon may be called coherent ionization, because of the trans-spectral coherency of all the photoelectron spectral components, which have the same initial phase; the result is
a well-ordered stratification of the photoelectron cloud. Based on Eq. (5), Fig. 2 displays a sequence of well-separated moving peaks in $|\Psi_{\text{ion}}(t,x)|^2$. The velocities of the respective electron bunches almost coincide with $\sqrt{2E_{\text{max}}/m}$, where $E_{\text{max}}$ are the energies of the local maxima of the photoelectron energy spectra $|b_\pm(E)|^2$, while the peak divides propagate with the velocities that correspond to minima in those spectra. Moreover, the relative heights of the peaks in $|\Psi_{\text{ion}}(t,x)|^2$ reflect the relative spectral amplitudes at the respective maxima. One can show that for weak kicks they are proportional to $(|E_0| + E)^{-4}E^{-1/2}$, $E_0$ being the ground-level energy.

Figure 2 also reflects the approximate backward-forward symmetry of the photoelectron cloud. The visual similarity of the emission parallel and antiparallel to the field is obvious. Quantitatively, the ratio of the electron emission along the force and in the opposite direction is just 1.2, with the total ionization probability for $q=1$ used quite substantial—$\sim 10^{-5}$. Even for $q=10$ (50% short of the kick that fully ionizes the well), for which the ionization probability is $\sim 7\%$ and the ground level is almost totally depopulated, the forward-backward ratio is still as small as $\sim 3$. As a result, the qualitative similarity remains, although one peak begins dominating the picture.

(ii) 3D hydrogen and hydrogenlike atom (HA). The amplitudes $b(k)$ of the $\delta$ ionization from the ground state can be extracted from the calculations on atomic collisions [11,12]; for an arbitrary initial state, see Ref. [13]. Based on Refs. [12,6], we write $\Psi_{\text{ion}}$ for the HA as follows (in atomic units):

$$
\Psi_{\text{ion}}(t,r) = \int dk \chi(k)\psi^-(k,r)\exp(-ik^2/t/2),
$$

$$
\psi^-(k,r) = (2\pi)^{-3/2} \Gamma(1+i/k)e^{\pi/2k}
\times e^{ik\cdot r} \frac{1}{1 + q^2 - k^2 - 2ik}
\times \frac{q^2 - q\cdot k(1 + ik^{-1})}{[1 + (q - k)^2]^2[1 + q^2 - k^2 - 2ik^2]},
$$

Here $\Gamma$ is the gamma function, and $\frac{1}{1 + q^2 - k^2}$ is the confluent hypergeometrical function. Substituting Eqs. (13) and (14) into Eq. (12), we derive $\Psi_{\text{ion}}$ as

$$
\Psi_{\text{ion}}(t,r) = \int dk \frac{4\exp(-ik^2/t/2)\exp(-i\gamma/k)}{\pi^2k[1 - \exp(-2\pi/k)]}\exp(-k^{-1}\tan^{-1}[2k/(1 + q^2 - k^2)])
\times \left[ q^2 - q\cdot k(1 + ik^{-1}) \right] 
\times [1 + (q - k)^2]^2(1 + q^2 - k^2 - 2ik^2)
\times \exp(ikr)\frac{1}{1 + q^2 - k^2}
\times \frac{1}{1 + q^2 - k^2} \frac{1}{1 + (q + k)^2} - \frac{1}{1 + (q - k)^2};
$$

the Kummer transformation of the confluent hypergeometric function was used in deriving Eqs. (15) to reduce the dependency on the direction of $r$. Integrated over energy, the angular distribution of photoelectrons based on Eq. (14) is presented in Ref. [7], and displays an apparent approximate “forward-backward” symmetry for a kick as strong as $q = 0.2$. The photoelectron spatial distribution based on Eq. (15) for a weak kick (Fig. 3) shows an apparent similarity with Fig. 2: an approximate symmetry with regard to the field direction and pronounced maxima (shells). Now, however, a shell does not necessarily correspond to electrons with close velocities, as is the case for a QW. Moreover, the spectra of $|b(k)|^2$ [Eq. (14)] do not contain any peaks for weak kicks. We believe that the shells in Fig. 2 are in fact due to the availability of ultrashort subcycle pulses. In this regard, one could argue that propagating subcycle EM pulses of what-
ever length are unphysical, as they contain a dc component. This argument, however, applies strictly only to the far-field area, while many experiments are feasible in the near-field area. Moreover, although known half-cycle pulses (HCP’s) [4] do consist of two parts of the opposite signs, practically all the ionization is due to the first, strong and short, part. Our computer simulations for a pulse fitted [10,14] to an experimental HCP confirm that we can safely neglect the HCP “tail.” Whether such pulses are δ like depends further on the ratio of the pulse duration to the time scale of the affected quantum system. For ionization, this time scale is determined by $E_{\text{ion}}$: $t_{\text{ion}} = \frac{\hbar}{E_{\text{ion}}}$. δ ionization of the ground-state hydrogen atom, therefore, would require a pulse duration of $\ll \approx 10^{-16}$ sec, which is beyond reach (but could become feasible in the future with the methods proposed in Ref. [2]). The effects considered here for the HA could, however, be observed in the ionization of Rydberg atoms, with $t_{\text{ion}}$ in picoseconds, by existing subpicosecond HCP’s, using, e.g., the visualization technique of Ref. [15] in combination with time gating. Similarly, relatively long pulses can emulate a δ kick for the ground-state ionization of a shallow QW. Moreover, our simulations for finite-width pulse ionization show many features of the δ ionization even for the pulses as long as $\hbar/U_0$. Therefore, with, e.g., $U_0 = 10$ meV, ~200-fs pulses will be short enough to observe the characteristic stratification; this duration is close to that of already available HCP’s. An almost δ-ionization-like picture would require pulses about four times shorter; even shorter pulses [2] will be needed for deeper QW’s. The evolving stratified photoelectron distribution might be observed using a combination of near-field microscopy with ultrafast time-resolved spectroscopy proposed recently for studying other spatiotemporally localized electronic phenomena in semiconductor QW’s (see, e.g., Ref. [16]). Since a particularly spectacular symmetry is predicted for a relatively weak kick and, consequently, a relatively low degree of ionization, one may employ additional techniques to improve the signal-to-noise ratio by, e.g., cooling the QW structure.

In conclusion, we have discovered theoretically that δ ionization of a one-dimensional QW by an ultrashort (δ-like) electromagnetic pulse would result in an ordered symmetrical stratification of the photoelectron cloud, as the trans-spectral coherence of the δ-like pulse maps the structure of the QW ionization continuum into photoelectron spatiotemporal distribution. A similarly ordered, symmetrical photoelectron cloud would result from δ ionization of a 3D hydrogen-like atom. The predicted effects may initially be observed experimentally in Rydberg atoms or in QW’s with a low binding potential.

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[8] D. K. Ferry, Quantum Mechanics IG (Institute of Physics and Physical Society, Bristol, 1995), Chap. 2.5.


