Discrete peaks in above-threshold double-ionization spectra

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Quantum-mechanical calculations of multiphoton double ionization by intense laser pulses show that the total-kinetic-energy spectrum of the photoelectrons consists of peaks separated by the photon energy, analogous to ordinary above-threshold ionization. Related structures appear in the two-electron and in the recoil-ion momentum distribution. We propose a method to extract the total-kinetic-energy spectrum from the experimental recoil-ion spectrum.

In this paper, we show numerically that laser pulses in the visible and ultraviolet produce distinct peaks in the double-ionization photoelectron spectra. Therefore, the experimental detection of such above-threshold double-ionization (ATDI) peaks should be possible. Even if we have experimental access only to the recoil-ion momentum spectrum, information about the peak structure in the full two-electron spectrum can be retrieved, as we will demonstrate below. This finding is highly nontrivial since the recoil-ion spectrum depends crucially on how the energy is shared between the electrons. The existence of ATDI peaks in the total-energy spectra is expected, but it is not obvious under which conditions they can be observed. In a very strong laser pulse, e.g., the atom is rapidly ionized within a few optical cycles. The formation of ATI or ATDI peaks, however, can be understood as interference between contributions ejected at different times over many cycles and therefore will not be observed for too short or too intense pulses.

The nonperturbative numerical solution of the full time-dependent two-electron Schrödinger equation in three dimensions (3D) [11] is very demanding and has been achieved so far only for small wavelengths, mainly in the ultraviolet range. The calculation of photoelectron energy spectra requires sufficiently dense continuum states (or, in other words, a large grid) and is therefore even more involved than the calculation of ionization probabilities. To demonstrate the existence of ATDI peaks, we therefore use the familiar one-dimensional model of a two-electron atom with soft Coulomb interactions [12], subject to an electric field \( E(t) \sin \omega t \). The time-dependent Schrödinger equation, in atomic units, for the two-electron wave function \( \Psi(z_1, z_2, t) \),

\[
\frac{\partial \Psi(z_1, z_2, t)}{\partial t} = \left[ \frac{p_1^2}{2} + \frac{p_2^2}{2} + (p_1 + p_2) A(t) \right] \Psi(z_1, z_2, t) = \left[ \frac{2}{\sqrt{z_1^2 + 1}} - \frac{2}{\sqrt{z_2^2 + 1}} + \frac{1}{(z_1 - z_2)^2 + 1} \right] \Psi(z_1, z_2, t),
\]

with \( A(t) = - \int_0^t E(t') \sin \omega t' \ dt' \), is solved numerically by means of the split-operator method [13]. As described in previous work [15], the configuration space spanned by the co-
due to the fact that the oscillating electric field shifts the 
shift towards lower energy with increasing intensity. This is 
different laser intensities are plotted. Apparently, the peaks 
photons energy.

peaks is constant throughout the spectrum and equals the 
stant energy. In each case, the separation between adjacent 
peaks that are in obvious correspondence to the rings of con-

momentum distributions. They consist of well-defined 
ionized electrons. At the end of the propagation, the 
wave function in the double-ionization region yields the mo-

mentum and energy distributions that we present in the fol-
lowing.

Figure 1 shows the two-electron momentum distributions 
and the total-kinetic-energy spectra after the action of laser 
pulses with a duration of 20 fs. The field is switched on and 
off using 5 fs linear ramps. The most striking features of the 
momentum distributions are concentric circles, which are 
most pronounced at short wavelength and low intensity.

Since the total kinetic energy is given by
\[ E_{\text{tot}} = p_1^2/2 + p_2^2/2, \]
these rings are lines of constant energy. The energy spectra 
the right-hand side of Fig. 1 are obtained by integration of 
these rings are lines of constant energy. The energy spectra

FIG. 1. Left: Two-electron momentum distributions for double 
ionization by laser pulses with wavelengths and intensities as 
indicated in the panels on the right-hand side. Right: Photoelectron 
total-kinetic-energy spectra for double ionization by laser pulses 
with wavelengths and intensities as indicated.

ordinates \( z_1, z_2 \) is divided into an inner region corresponding 
to the neutral atom (\( |z_1|, |z_2| < a \)) and two outer regions 
corresponding to single ionization (\( |z_1| < a, |z_2| \geq a \) or \( |z_1| \geq a, |z_2| < a \)) and double ionization (\( |z_1|, |z_2| \geq a \)), respectively. In the outer parts, the electron-electron interaction is 
 neglected, as well as the interaction between the nucleus and 
the far-away electrons. At the end of the propagation, the 
wave function in the double-ionization region yields the mo-
mementum and energy distributions that we present in the fol-
lowing.

Figure 2 shows the two-electron momentum distributions 
and the total-kinetic-energy spectra after the action of laser 
pulses with a duration of 20 fs. The field is switched on and 
off using 5 fs linear ramps. The most striking features of the 
momentum distributions are concentric circles, which are 
most pronounced at short wavelength and low intensity.

Since the total kinetic energy is given by
\[ E_{\text{tot}} = p_1^2/2 + p_2^2/2, \]
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these rings are lines of constant energy. The energy spectra

energy of the continuum states and thereby increases the ion-
ization barrier. Analogous to ordinary short-pulse ATI, the 
position of the peaks can be approximately calculated as

\[ E_n = n \hbar \omega - (I_p^{(2)} + 2 U_p), \]
where \( n \) is the number of absorbed photons, \( I_p^{(2)} \) is the 
double-ionization potential, and \( U_p = E^2/(4 \omega^2) \) is the ponderomotive potential (which we evaluate at the peak ampli-
tude). The increase in the ionization threshold amounts to \( U_p \) 
per electron. Hence the term \( I_p^{(2)} + 2 U_p \) appears, as opposed 
to ordinary ATI, where we have \( I_p^{(1)} + U_p \). In Eq. (2), it is 
assumed that the ac Stark shift of the ground state is negligi-
ble. The energies predicted by Eq. (2) are indicated by 
arrrows in Fig. 2; they give a good estimate for the peak 
energies. The number of absorbed photons is specified for 
each arrow. Using Floquet theory [14], we have calculated 
the quasi-ground-state energies and have confirmed that the 
small discrepancy is indeed due to the ac shift of the ground 
state: The ac shifts corresponding to the laser parameters of 
Fig. 2 are 0.0096 a.u. for \( 2 \times 10^{14} \) W/cm\(^2\), 0.014 a.u. for 
\( 3 \times 10^{14} \) W/cm\(^2\), and 0.025 a.u. for \( 4 \times 10^{14} \) W/cm\(^2\). The 
differences between the numerical peak energies and Eq. (2) 
are, on average, 0.009 a.u., 0.015 a.u., and 0.020 a.u., respec-
tively. The very small remaining discrepancies are due to 
contributions from the leading and falling edge of the pulse 
where the intensity is below its peak value, or due to small 
umerical errors. The substructures in Fig. 2 come from 
laser-induced resonances giving rise to an additional series of 
peaks, also separated by the photon energy. This can be con-
firmed by inspection of the corresponding Floquet spectra.

We now turn our attention to the distribution of recoil-ion 
momenta (Fig. 3), because these can be measured with much 
less effort than the full correlated two-electron spectra [16]. 
As \( P = -(p_1 + p_2) \), the recoil-ion momentum spectrum is 
calculated by integration of the two-electron momentum dis-
tribution. Again, we find a pronounced peak structure, at 
least in the case of small wavelength and intensity. However,
the circle. In this case, we have the integral as largest when the integration is along a tangent to the lower and upper ends, because the value of the vertical line integral is the same as the upper part of Fig. 4 shown as the lower part of Fig. 4. A single ring with energy \( E_{\text{tot}} \) in the \( (p_1, p_2) \) plane gives rise to a distribution over \( p \) with two peaks and sharp cutoffs at the lower and upper ends, because the value of the vertical line integral is largest when the integration is along a tangent to the circle. In this case, we have \( p_1 = p_2 = \pm \sqrt{E_{\text{tot}}} \). Consequently, the peaks are located at \( \pm P_{\text{max}} = \pm 2\sqrt{E_{\text{tot}}} \), in agreement with the above result. The structures in the inner region of the \( P \) distributions originate from structures within the rings, mainly within the innermost ring. For 400 nm and 10^{15} \text{ W/cm}^2, the calculation does not resolve the ATDI peaks even though the spacing must be the same as in the case of 3 \times 10^{14} \text{ W/cm}^2. The reason is twofold: (i) The outer ATDI rings (left-hand side of Fig. 1) have a more pronounced substructure, and (ii) the peaks in the energy spectrum (right-hand side of Fig. 1) are broadened due to pulse-shape effects (contributions of various intensities while the laser is switched on and off.)

In general, the electron total-kinetic-energy spectrum cannot be retrieved from the recoil-ion spectrum because information is lost in the integration procedure. However, under some reasonable assumptions, such a reconstruction should be possible. To that end, we introduce a function \( h(E, \phi) \) that is the distribution of the total energy \( E \) and the “angle” \( \phi \), which is defined by \( \tan \phi = p_2 / p_1 \). Under the assumption that \( h(E, \phi) \) is a product of an energy distribution and an angular distribution, \( h(E, \phi) = h_E(E) h_\phi(\phi) \), and that the spectra have forward/backward symmetry \( h_\phi(\phi) = h_\phi(3\pi/2 - \phi) \), we calculate the recoil-ion momentum distribution \( R(P) \) as

\[
R(P) = \int_{-P/2}^{P/2} h_E(E) h_\phi \left( \arctan \frac{P + 2f(E,P)}{P - 2f(E,P)} \right) \frac{dE}{f(E,P)},
\]

where \( f(E,P) = \sqrt{E - (P^2/4)} \). We assume further that the energy spectrum does not extend up to infinity so that the upper limit of the integral can be replaced by a finite value \( E_{\text{max}} \). Then, for given functions \( R(P) \) and \( h_\phi(\phi) \), Eq. (3) is a Volterra integral equation for \( h_E(E) \) and can readily be solved numerically [17]. Figure 1 shows that the two-electron distributions are not very anisotropic in the \( (p_1, p_2) \) plane. (This is different for long wavelengths around 800 nm [9,15].) Therefore, we simply use a constant function \( h_\phi \)
shows the exact calculated energy spectrum, and panel distribution of 0.04 a.u. oscillations can be suppressed by convolution with a Gaussian. Thus we arrive at the spectrum in panel ~h~. The particular choice of 3 nm pulse with intensity 3 \times 10^{14} \text{W/cm}^2. The upper panel shows the exact calculated energy spectrum, and panel (b) shows the spectrum that is reconstructed from the recoil-ion momentum distribution by the solution of Eq. (3). We find peaks at the correct positions, but also we find a rapidly oscillating substructure and negative values of h_E. The oscillations can be suppressed by convolution with a Gaussian distribution of 0.04 a.u. width (full width at half maximum). Thus we arrive at the spectrum in panel (c). Considering the crudeness of the approximation, the agreement with the exact spectrum is excellent: The peak positions are correct, and the envelope is well reproduced except for small energies lower than 0.5 a.u.

Equation (3) has been derived for the 1D model, knowing that the photoelectrons are preferentially ejected along the laser polarization: Ref. [18] indicates that the angle between the two electrons deviates not more than \pi/6 from zero or \pi. Together with the fact that the recoil-ion momentum is strongly aligned with the polarization axis [8,19], we conclude that for nearly equal moduli of the electron momenta, the angle of electron emission is typically less than \pi/12, reducing the peak positions in the recoil-ion spectra by at most a factor of \cos(\pi/12) \approx 0.97. Thus we expect that the reconstruction procedure can be applied to experimental spectra and will be helpful in the detection of the ATDI peaks.

To summarize, we have demonstrated the existence of ATDI peaks for a variety of laser parameters. The peaks appear not only in the photoelectron total-energy spectra, but also in the recoil-ion momentum distributions, which can be measured much more easily. We have proposed a numerical method to approximately extract the former from the latter.

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