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Channel-closing-induced resonances in the above-threshold ionization plateau

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Multiphoton resonances with ponderomotively upshifted Rydberg states are believed to have dramatic effects on the plateau of high-order above-threshold ionization. For short pulses and intense laser fields, individual Rydberg states lose their physical significance. Under these conditions, on the basis of experimental and theoretical investigation of the dependence of the photoelectron spectra on the laser intensity, we propose that the observed effects can be largely understood in terms of the channel closings that are characteristic of a short-range model potential.

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Above-threshold ionization (ATI) is photoionization of atoms in strong laser fields [1]; for a review see Ref. [2]. In the present paper we investigate a phenomenon that has recently caught significant interest: strongly intensity-dependent enhancements of groups of ATI peaks that have been attributed to resonance with ponderomotively upshifted bound states close to the ionization limit (Rydberg states) [3–5]. We describe experiments performed with the highest electron count rates and the shortest laser pulses (50 fs) used in this kind of work. With such intense and short pulses, the excited bound states of the atoms under investigation are broadened so dramatically that their significance is put into question. The features of the spectra presented here are studied in a Keldysh-type theoretical approach that does not incorporate any excited bound states, and explained by channel closings.

We start by reviewing the key points of the present understanding of ATI. For fixed laser intensity, electrons are emitted with energies

$$E_n \equiv \mathbf{p}_n^2/(2m) = n\hbar\omega - |E_0| - U_p, \tag{1}$$

with E_0 the ground-state energy of the atom, ω the laser frequency, and U_p the ponderomotive energy, i.e., the cycle-averaged kinetic energy of a free electron in the presence of the laser field. The vector $\mathbf{p}_n$ specifies the electron's momentum at the detector, i.e., its drift momentum. The envelope of the electron's spectrum exhibits various features that are independent of the atomic species: (i) ionization such that the ionized electron never re-encounters the ion yields a maximal classical energy of $2U_p$ [6]; (ii) enhanced ionization occurs for intensities where a ponderomotively upshifted Rydberg state is multiphoton resonant with the ground state (Freeman resonance [7]); (iii) an ionized electron that does re-encounter the ion and elastically rescatters may acquire a maximal classical energy of $10U_p$ [8,9]; (iv) different electronic "quantum trajectories" may interfere, for a fixed laser intensity, constructively or destructively, and under auspicious circumstances these interferences remain visible in experimental spectra [10,11].

In the past few years, new data [3,4] gave evidence of some resonance-like mechanism that is able to enhance the yields of groups of ATI peaks within the lower two thirds of the plateau by up to one order of magnitude at certain precisely defined intensities. These "resonances" are well reproduced by simulations using the single-active-electron approximation [12–14]. In fact, the simulations suggest that they too are manifestations of the Freeman-resonance mechanism, surprisingly strong and for surprisingly high electron energies. Inspection of the time evolution of the wave function has shown [5] that the resonances are related to electrons that linger around the ion for several cycles of the field before they finally escape.

The experiments discussed above [3,4,12] employed comparatively long laser pulses exceeding 100 fs. The present paper deals with the questions of what happens to these resonances for short pulses, and how they are afforded by a simple model of ATI. Notice that our pulse duration is shorter than the classical revolution times of the higher Rydberg states.

We will show experimental data that prove the persistence of the "resonant" enhancements and results of a theoretical model [15] that qualitatively reproduce the experiments and allow us to relate the resonant intensities to "channel closings" [16]. As the model potential, we will use the zero-range potential that supports one single bound state (with the binding energy $E_{0,\mathrm{zr}}$). All continuum states are upshifted by the ponderomotive energy U_p. Whenever the upshifted continuum threshold is multiphoton resonant with the ground state, viz.

$$|E_{0,\mathrm{zr}}| + U_p = k\hbar\omega \tag{2}$$

for some integer k, we speak of a channel closing. In this case, owing to Eq. (1), an ionized electron (in its final state or in its intermediate state prior to rescattering) can have an effective drift momentum of zero (for $n=k$).

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An electron bound to a *real* atom experiences the tail of the Coulomb potential that causes a Rydberg series below the continuum threshold. However, in experiments with short laser pulses, the higher Rydberg states cannot be resolved, owing to the energy-time uncertainty constraints or their field-induced Floquet widths. It makes good sense to introduce some bona fide continuum threshold at an energy $-|E_c|$ such that states with energies higher than that behave, for the purposes of interest, like continuum states [17]. We then treat the real atom as one bound electron characterized by two parameters: its binding energy E_0 (which, via energy conservation, determines the energies (1) of the ATI peaks) and the continuum threshold (which is relevant for the channel closings). In doing so, we will make the identification $|E_{0,\mathrm{zr}}|=|E_0|-|E_c|$.

Qualitatively, the effects we will discuss do not depend on the choice of E_c. However, for semiquantitative agreement between experiment and theory, the choice of E_c can be crucial. Various criteria can be invoked to establish a specific value of E_c [18]. Here, we will proceed as follows: the field-free energy-level density of any rare-gas atom is very uneven, showing noticeable gaps at certain energies. We will use the upper end of the gap closest to the continuum to define E_c. We assume that all excited states in question suffer the same ponderomotive shift as the continuum. In place of the condition (2), the channel closing in the real atom is now defined by $|E_0|-|E_c|+U_p=k\omega$.

Our experimental analysis is based on a careful investigation of the intensity dependence of the ATI spectra. The most important issue for investigating effects in the plateau region is the ability to record as many photoelectrons as possible within a reasonable measurement time. By using a femtosecond laser system with a repetition rate of 100 kHz, we are able to record more than 300 million electrons in a few hours even though we vary the laser intensity. The pulse duration is about 50 fs, the pulse energy 6 μJ, and the wavelength 800 nm. Peak intensities in excess of 10^{14} W/cm^2 can be achieved. A half-wave plate mounted on a rotary stage and followed by a linear polarizer is used in order to fine-tune the intensity of the laser pulses, while coarse tuning is done by changing the focusing geometry. All the measurements presented here are for linear polarization in the direction towards the electron detector. Spectra in an intensity interval of $(0.3–1.0)I_0$ in steps of $0.01I_0$ were recorded where I_0 is the maximum intensity for the corresponding measurement and depends on the focusing geometry.

Throughout this paper, argon is investigated, and as discussed above, we will envision it as a one-electron bound system with the continuum threshold at -1.07 eV. This is where the spectrum of argon exhibits a pronounced gap, situated near the $4f$ states. The following intensities of channel closings ensue: 1.36 ($k=10$), 3.97 ($k=11$), 6.57 ($k=12$), 9.17 ($k=13$), and 11.77 ($k=14$) in multiples of 10^{13} W/cm^2. The lowest two do not have noticeable consequences within the intensity range that we consider, the last three will be instrumental for the discussion.

Figure 1(a) shows a few examples of ATI spectra that were measured while recording an intensity distribution as just described with $I_0\approx 8\times 10^{13}$ W/cm^2. In general, the spectra behave as expected: As the intensity increases, the counts rise. In addition, the ATI plateau develops. A closer look at the spectra reveals a striking difference between the spectra for $I\leqslant 0.8I_0$ and those for higher intensity: Within a small intensity interval, a group of ATI peaks corresponding to energies between about 15 eV and 25 eV grows very quickly. In the figure this is emphasized by horizontal lines drawn at the maximal heights of the plateaus. Increasing the intensity above $0.9I_0$ leads to smaller growth rates of those peaks. The plateau, however, preserves its shape. For an interpretation, it should be kept in mind that a measured ATI spectrum is made up by contributions from all intensities $I\leqslant I_0$ that are contained within the spatio-temporal pulse profile. This means that a spectrum for a fixed intensity would show the enhanced group of ATI peaks only at that intensity where it first appears in our measurement, namely at $I\approx 0.85I_0=7\times 10^{13}$ W/cm^2, just above the $k=12$ channel closing. In other words, the enhancement happens at a well-defined intensity or at least within a very small intensity interval. Two additional features of Fig. 1(a) are important: First, the apparent cutoff of the spectrum, defined to be slightly after the energy where the spectrum starts its final roll-off, makes a dramatic jump when the intensity rises from 0.8 I_0 to 0.9 I_0. Second, at the channel closing, the contrast of the spectrum appears to be reduced, in agreement with a related recent experiment [19].

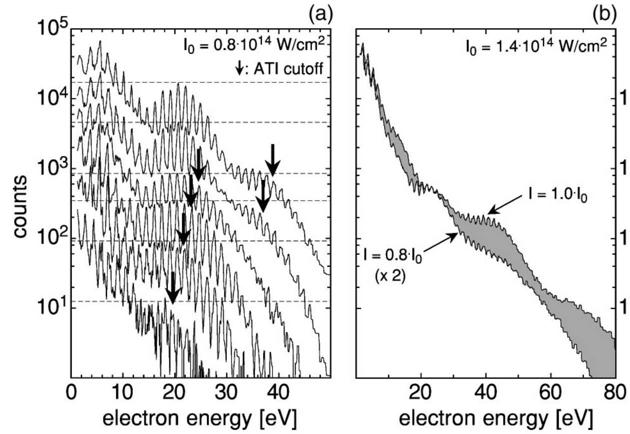

FIG. 1. (a) ATI spectra in argon at 800 nm recorded in the direction of the linearly polarized field for various intensities equally distributed between $0.5I_0$ and $1.0I_0$ with $I_0\approx 8\times 10^{13}$ W/cm^2. The horizontal lines mark the maxima of the plateaus for each intensity. The apparent cutoff position of the spectra is indicated by an arrow. (b) Same as (a), but for $I_0=1.4\times 10^{14}$ W/cm^2. The two traces are for $1.0I_0$ and $0.8I_0$, the counts in the latter case have been multiplied by a factor of 2.

The results of modeling the data of Fig. 1(a) are shown in Fig. 2. One notices how a group of peaks rears up at the intensity of the channel closing [20]. At the same intensity, the final group of peaks that precede the cutoff is suppressed. Hence, when the intensity increases through the channel closing, the apparent cutoff will experience the kind of jump observed in the data. The two insets in Fig. 2 show the approximation of the exact calculations in terms of ‘‘quantum trajectories’’ [21], below and right at the channel closing. In each case, we present the spectra resulting from the

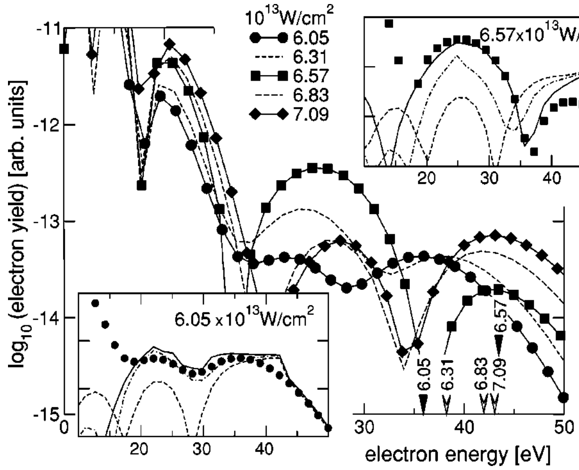

FIG. 2. Calculated ATI spectra corresponding to Fig. 1(a) for five different intensities, as indicated in W/cm^2 in the figure. The central intensity is at the position of the channel closing with multiphoton order $k=12$ ($I=6.57\times10^{13}$ W/cm^2). For each intensity, the position of the final maximum of the spectrum (the cutoff) is marked by an arrow. The two insets illustrate the approximation of the calculation in terms of quantum trajectories. The inset at the upper right is for the $k=12$ channel-closing intensity, the one at the lower left for the lower intensity of 6.05×10^{13} W/cm^2. The depicted approximations include the first three pairs of orbits (dashed line), the first ten pairs (dot-dashed line), and the first twenty pairs (solid line). Only rescattering orbits are included; therefore, the approximations do not work for energies below 20 eV.

first 3, 10, and 20 pairs of trajectories (ordered according to the length of their travel time). Below the channel closing, ten pairs already provide an excellent approximation (in fact, already six do). This implies that the contributions of the longer travel times add up in a random fashion. In contrast, at the channel closing the first ten pairs are still off by almost half an order of magnitude. The exact result is generated by constructive interference of quantum trajectories with exceptionally long travel times. (The travel times of the two trajectories of the nth pair are approximately $n/2$ optical cycles.) In close correspondence, it was noticed in the simulations [5,13,14] that the electrons that produce the resonant enhancements dwell near the nucleus for many cycles before they finally rescatter.

For the zero-range channel closings, a precondition for the constructive interference is the occurrence of many recollisions, which is made possible by the electron having zero drift momentum at a channel closing. For the real atom, for the electron to have zero drift momentum outside the range of the potential (i.e., at infinity), it will have to escape quickly from the vicinity of the ion. Indeed, the simulations [13,14] show no resonance at the continuum-threshold channel closings given by $|E_0|+U_p=k\hbar\omega$, but rather at $|E_0|-|E_c|+U_p=k\hbar\omega$, where the electron can be trapped in a wave packet of laser-dressed highly excited bound states [23].

Only very few quantum trajectories contribute to the upper third of the ATI rescattering plateau [22]. As a consequence, the conspiracy that is responsible for the enhancements at the channel closings can only efficiently succeed in

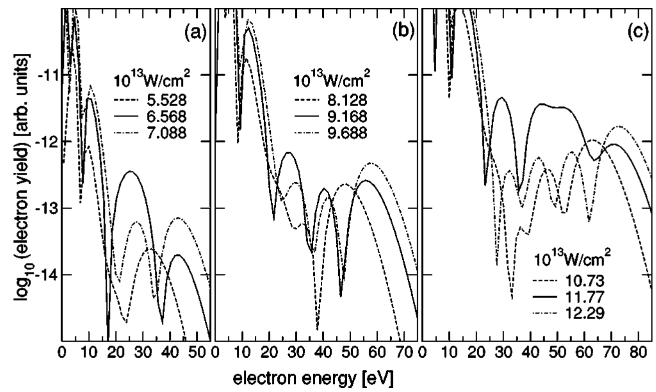

FIG. 3. The panels (a) to (c) display the envelopes of calculated ATI spectra at, below, and above the intensities of the 12, 13, and 14-photon channel closings as indicated in the panels.

the lower two thirds of the plateau. This agrees with all experimental observations and with numerical simulations [5,13,24]. The decrease of the highest peaks just below the cutoff that parallels the enhancements at the channel closings is not in conflict with this statement: it is substantially less pronounced than the enhancement.

Further support for this interpretation comes from Fig. 1(b) and Fig. 3, which display data and respective results of the theoretical model along the lines of Fig. 2, but for a higher maximal intensity I_0 so that more than one channel closing plays a role within the intensity profile of the pulse. For the data, by evaluating the energy position of the ATI plateau cutoff as well as by comparing the spectra to those of Fig. 1(a), I_0 is estimated at 1.4×10^{14} W/cm^2. Owing to the higher maximum, the enhancement described above, involving the peaks with energies between 20 and 25 eV, now already appears at $0.5I_0$. Increasing the intensity leads to more and more electrons with high energy, while the appearance of the spectra qualitatively remains the same. At an intensity of $I\approx0.85I_0=1.2\times10^{14}$ W/cm^2, again a group of ATI peaks begins to grow quickly within a very small intensity interval. This group of peaks is located at energies between about 35 eV and 45 eV and its height finally approaches the height of the group of ATI peaks described above. The two intensities just mentioned are very close to the $k=12$ and $k=14$ channel closing intensities. No effect of comparable clarity could be observed at the intensity corresponding to the $k=13$ channel closing. In Fig. 3 we compare these data to the results of the same theory as above. In each case, at the channel closings we observe a marked enhancement of the group of peaks between 20 and 30 eV, strong for $k=12$ and $k=14$, weak for $k=13$. This agrees with the data and was also observed in numerical simulations [13]. For the highest intensity, in addition the group of peaks between 40 and 55 eV is enhanced by up to one order of magnitude.

In conclusion, we have presented data showing that the enhancements of the ATI rescattering plateau persist for short laser pulses. We have made the case that whenever the Rydberg series loses its significance, owing to strong fields or short pulses, then the data can be understood and qualitatively described by an idealized atom having just one bound state and an effective continuum starting at about the first

Rydberg state of the real atom that can no longer be resolved. The conceptual simplification achieved by this model is the replacement of the hard-to-envision field-perturbed Rydberg orbits [14] by the comparatively simple quantum orbits [21], which are essentially the orbits of the so-called simple-man model [6,9]. Near the channel closings, a large number of orbits conspire to produce a constructive interference that raises groups of peaks in the lower two thirds of the rescattering plateau by up to one order of magnitude. Hence, for sufficiently short pulses and intense fields, we may add a fifth mechanism to the four mentioned in the introduction: (v) strong enhancements within the lower two thirds of the plateau occur at intensities corresponding to the channel closings defined above. In the upper third of the plateau, the shortest two orbits are very dominant so that interference effects are less pronounced. Still, some interference does occur, and it is destructive.

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[1] P. Agostini *et al.*, Phys. Rev. Lett. **42**, 1127 (1979).
[2] C. J. Joachain *et al.*, Adv. At., Mol., Opt. Phys. **42**, 281 (2000).
[3] M. P. Hertlein *et al.*, J. Phys. B **30**, L197 (1997).
[4] P. Hansch *et al.*, Phys. Rev. A **55**, R2535 (1997).
[5] H. G. Muller and F. C. Kooiman, Phys. Rev. Lett. **81**, 1207 (1998).
[6] H. B. van Linden, vanden Heuvell, and H. G. Muller, in *Multiphoton Processes*, edited by S. J. Smith and P. L. Knight (Cambridge University Press, Cambridge, 1988).
[7] R. R. Freeman *et al.*, Phys. Rev. Lett. **59**, 1092 (1987).
[8] G. G. Paulus *et al.*, Phys. Rev. Lett. **72**, 2851 (1994); G. G. Paulus *et al.*, Phys. Rev. A **52**, 4043 (1995).
[9] P. B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).
[10] R. Kopold and W. Becker, J. Phys. B **32**, L419 (1999).
[11] G. G. Paulus *et al.*, Phys. Rev. Lett. **80**, 484 (1998); **84**, 3791 (2000).
[12] M. Nandor *et al.*, Phys. Rev. A **60**, R1771 (1999).
[13] H. G. Muller, Phys. Rev. A **60**, 1341 (1999).
[14] H. G. Muller, Opt. Express **8**, 44 (2001).
[15] A. Lohr *et al.*, Phys. Rev. A **55**, R4003 (1997). This builds on the strong-field approximation, as introduced by L. V. Keldysh, Zh. Éksp. Teor. Fiz. **47**, 1945 (1964) [Sov. Phys. JETP **20**, 1307 (1965)]; F. H. M. Faisal, J. Phys. B **6**, L89 (1973); and H. R. Reiss, Phys. Rev. A **22**, 1786 (1980), but includes rescattering.
[16] Channel closings in the *total* multiphoton detachment rate of H^- have been observed by C. Y. Tang *et al.*, Phys. Rev. Lett. **66**, 3124 (1991); for calculations see W. Becker *et al.*, Phys. Rev. A **42**, 4416 (1990); and W. G. Greenwood and J. H. Eberly, *ibid.* **43**, 525 (1991); for the effect on the spectrum, see H. M. Faisal and P. Scanzano, Phys. Rev. Lett. **68**, 2909 (1992). None of these references deals with the ATI plateau.
[17] L. D. Landau and E. M. Lifshitz, *Relativistic Quantum Mechanics* (Pergamon, Oxford, 1974), Chap. 90.
[18] M. Yu. Ivanov *et al.*, Phys. Rev. A **54**, 742 (1996).
[19] E. Cormier *et al.*, J. Phys. B **34**, L9 (2001).
[20] The theoretical results presented here are not averaged over the laser's intensity profile. Doing so produces spectra in close agreement to the experiment (these will be presented elsewhere), but obscures to some extent the underlying physics.
[21] Quantum trajectories have been introduced in order to analyze high-harmonic generation [M. Lewenstein *et al.*, Phys. Rev. A **49**, 2117 (1994)] and ATI [M. Lewenstein *et al.*, *ibid.* **51**, 1495 (1995)] in terms of the orbits of an electron that is ionized at some time t_0 by the laser field, subsequently propagates in its presence, and finally returns at some later time t_1 to the core where it recombines or rescatters. In most cases, the first four or six orbits, those having the shortest travel times t_1-t_0, provide an excellent approximation. The orbits are complex owing to their origin by tunneling. Their contributions interfere, which is responsible for the complex structure of high-harmonic generation and ATI spectra. The orbits constitute the quantum version of the simple-man model [6,9]. For a detailed account, see Ref. [22].
[22] R. Kopold *et al.*, Phys. Rev. Lett. **84**, 3831 (2000), for elliptical polarization and Opt. Commun. **179**, 39 (2000) for linear polarization.
[23] A graph of a pertinent orbit can be found in Fig. 11 of G. L. Yudin and M. Yu. Ivanov, Phys. Rev. A **63**, 033404 (2001).
[24] H. G. Muller, Phys. Rev. Lett. **83**, 3158 (1999).