Plateau in Above Threshold Ionization Spectra

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We present photoelectron energy spectra for the rare gas atoms in strong 40 fs, 630 nm laser pulses. A new property in the above threshold ionization distribution is described, namely, a plateau. Numerical calculations using one- and three-dimensional models suggest that at least in part this is a one-electron effect. All rare gas atoms investigated show similar behavior, indicating that the plateau in above threshold ionization is a universal phenomenon. We discuss a simple mechanism possibly responsible for the plateau.

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It is becoming increasingly evident that above threshold ionization (ATI) in the high intensity, nonperturbative regime is considerably more complex than has been assumed until now. Its companion phenomenon high order harmonic generation (HOHG) [1] appears to have settled into a reasonably well understood pattern, exhibiting a plateau and an abrupt cutoff. Numerical simulations for realistic atomic models have produced very good agreement with experimental data [2], have provided quantitative predictions for the range of the plateau [3], and are generally compatible with alternative interpretations [4–6]. A basic feature of HOHG is that it represents a phenomenon due to one active electron driven by the strong field and scattering the photons in the vicinity of the effective potential it feels whenever it passes near its origin. Effects involving the excitation and/or ionization of one or more additional electrons would destroy or severely alter the harmonics.

ATI on the other hand had until recently been assumed to exhibit the well known sequence of decreasing peak heights in the photoelectron energy spectrum over about 4 orders of magnitude of the intensity [7,8], with the corresponding angular distributions becoming increasingly peaked along the direction of the polarization vector (for light linearly polarized). Substructures [9,10] in the photoelectron energy peaks due to bound states shifting into resonance do not represent a departure from this basic picture and have been given the proper interpretation. A first indication that the situation may be more complex came from very recent experimental data [11] showing the presence of side lobes in the photoelectron angular distributions above certain photoelectron energy. Nonperturbative time-dependent calculations [11] have also produced such lobes at the experimentally correct energy range.

It is the purpose of this paper to present results that provide surprising evidence for a strong departure from the standard picture suggesting a hitherto unsuspected layer of effects. In short, ATI photoelectron energy spectra, obtained with 40 fs, 630 nm pulses in all of the rare gases, at intensities up to 4.4×10^{14} W/cm^2, exhibit an unexpected change of the slope of the envelope of the peak heights above about 20 eV. As we move from He to Xe this change of slope becomes increasingly pronounced to the point that in Xe (as well as in Ar) a clear plateau develops at a certain range of intensities. It should be noted here that the peak integrated photoelectron signal in Fig. 4 of Ref. [7] showed slight modulations between 8 and 25 eV. However, since the corresponding ATI spectra (Fig. 3 of Ref. [7]) decreased smoothly, the origin and meaning of those modulations remain uncertain, although in the light of our results it is conceivable that under different experimental conditions they might exhibit a marked change of slope.

The experimental setup [12] consists of a colliding pulse mode-locked oscillator which is amplified in a two-stage dye amplifier pumped by a copper vapor laser at a repetition rate of 6.2 kHz. This system delivers a maximum output energy of 20 μJ per pulse and after compression in a prism sequence, a pulse duration below 40 fs at 630 nm. The beam is focused inside the interaction chamber giving rise to intensities up to about 5×10^{14} W/cm^2. A standard time-of-flight spectrometer together with suitable fast electronics and a PC is used to analyze the photoelectrons. By means of a cryopump the background pressure was held below 10^{-8} torr. The target gas pressure in the chamber was 10^{-7} to 10^{-4} torr. The high repetition rate together with the extremely short pulse duration allows recording ATI spectra with high sensitivity, as we have also demonstrated in earlier work [12] on ATI in Cs.

Figure 1 shows measured ATI spectra for the five rare gases at an intensity of 3×10^{14} W/cm^2 (for He) and 2×10^{14} W/cm^2 (for the others) all of which exhibit either a sudden change of slope or a plateau. The appearance and evolution of this effect with increasing intensity in Ar is shown are Fig. 2. The overall evolution is typical of all five rare gases, although under the conditions of our

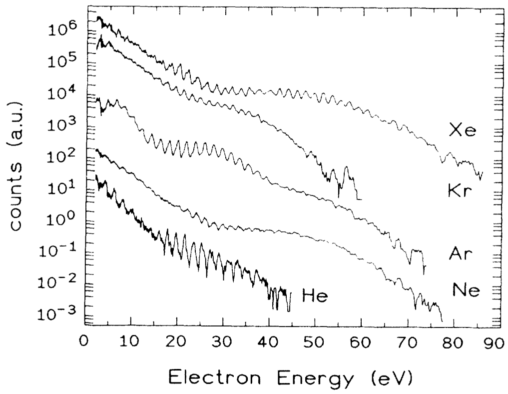

FIG. 1. ATI spectra of all rare gases. The intensity was 3×10^{14} W/cm^2 for He and 2×10^{14} W/cm^2 for all the others. (The curves are separated in the vertical direction.)

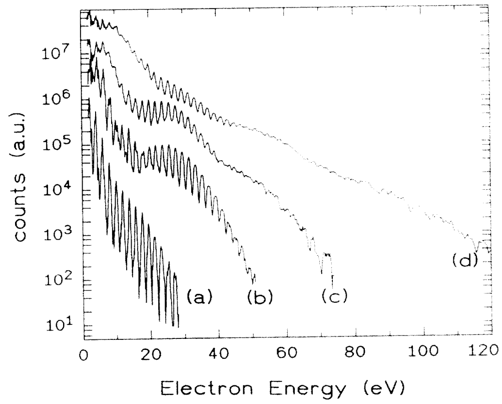

FIG. 2. ATI spectra from Ar with 40 fs, 630 nm pulses at intensities of 6×10^{13} W/cm^2 (*a*), 1.2×10^{14} W/cm^2 (*b*), 2.4×10^{14} W/cm^2 (*c*), and 4.4×10^{14} W/cm^2 (*d*) (the curves are separated slightly in the vertical direction for visual convenience).

experiment it is most pronounced in Ar and Xe. If we trace the development of the low energy photoelectron peaks from lower to high intensity, the onset of tunnel ionization becomes evident as the peaks tend to broaden and eventually smear out. This is compatible with the fact that for Ar, for example, at an intensity of 2×10^{14} W/cm^2, the Keldysh parameter $\gamma=\sqrt{E_{\mathrm{IP}}/2U_P}$ (where E_{IP} is the ionization potential of the bare atom and U_P the ponderomotive energy) is equal to unity. At the same intensity, however, the ATI peaks at the plateau are clearly resolved and at moderate field intensities, the plateau's envelope exhibits a maximum which, with increasing intensity, reduces to a change of slope.

A few significant aspects should be noted here: (a) The plateau cannot be attributed to peak suppression due to channel closing. At 2×10^{14} W/cm^2, for example, the ponderomotive shift of the ionization potential is only 4.4 eV, which means that at most the first two or three peaks may be suppressed, whereas the plateau begins with approximately the tenth peak. (b) Examination of the energy difference between a peak on the plateau and one at lower energies reveals that it does not correspond to an integral number of photons. This may be viewed as an indication that an additional mechanism is contributing to the formation of the plateau. (c) Measurements with light circularly polarized have exhibited neither a plateau nor a change of slope (Fig. 3).

As a first attempt to obtain the simplest possible indication of such a plateau in a theoretical model, we have performed a numerical simulation using a one-dimensional (1D) one-electron model with the potential $V(x)=-1/\sqrt{1+x^2}$ first employed in ATI calculations by Javanainen and co-workers [13,14]. We have solved the 1D time-dependent (TD) Schrödinger equation by expanding the wave function in terms of *B* splines in a finite element approach [15,16], which is different from that of Refs. [13] and [14]. A sample of results of such a calculation is shown in Fig. 4. Although we have used parameters similar to those of Refs. [13] and [14] our results are substantially different, possibly due to the smallness of the numerical grid employed in those calculations, especially if the authors had not anticipated significant presence of high energy electrons. For the set of parameters (photon frequency) $\omega=0.07$ a.u. (peak field strength), $\varepsilon=0.04$, 0.06, 0.08 a.u., total pulse length of 20 cycles (with a sine-square pulse envelope) we obtain an ATI spectrum that exhibits the qualitative behavior of the measured spectra in Ar and Xe at comparable intensity. Keeping in mind the limitations of a 1D model, the point to be made by this calculation is that the plateau seems to be, at least in part, a one-electron effect. As always, one expects effects to be somewhat exaggerated in a 1D model. We have, however, also obtained results through a 3D time-dependent calculation in hydrogen using the method and programs employed in previous work [17]. One result is shown in Fig. 5. It resembles the data

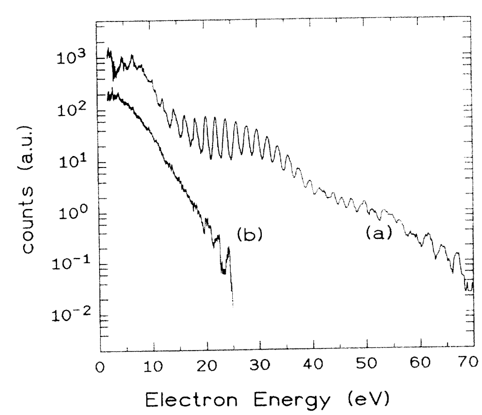

FIG. 3. ATI spectra from Ar at an intensity of 2.4×10^{14} W/cm^2 for linearly (*a*) and circularly (*b*) polarized light.

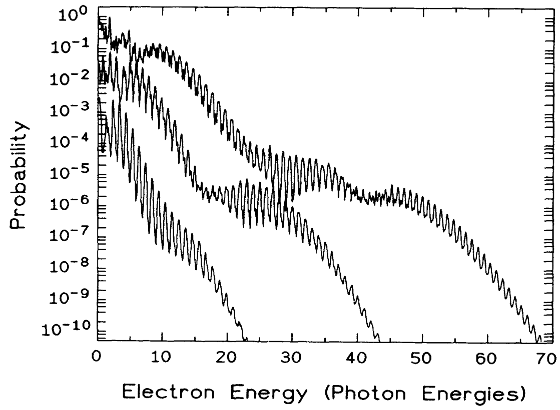

FIG. 4. ATI spectra obtained from a numerical simulation with a one-dimensional model atom. The frequency corresponds to 2 eV, the total pulse length was 20 cycles, and the maximum intensities were 5.6×10^{13} W/cm^2, 1.3×10^{14} W/cm^2, and 2.2×10^{14} W/cm^2.

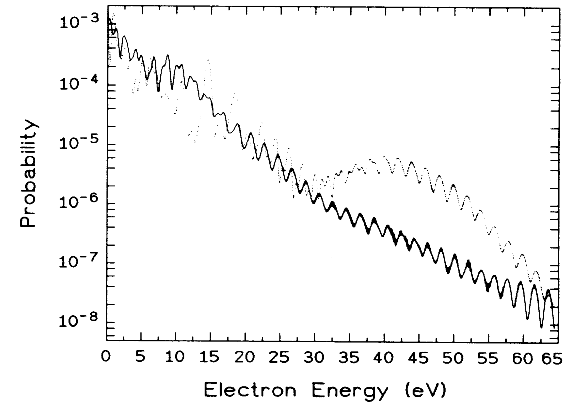

FIG. 5. ATI spectrum from a 3D calculation in H at an intensity of 2×10^{14} W/cm^2, pulse length 18 cycles, with a Gaussian shape and photon energy 2 eV. The dotted line has been obtained with a simplified model involving only three angular momenta which is responsible for the pronounced maximum resembling the behavior of the 1D model.

in He showing a change in slope but not a plateau. This is reasonable since the dipole matrix elements (i.e., the coupling constants) in H are significantly smaller than in Ar, Kr, and Xe although the ionization potentials are comparable.

Having established that a TD calculation predicts this effect, one should speculate about some underlying physical picture as well as about additional mechanisms, since it may not be true that all rare gases behave like one-electron systems under these circumstances.

It has been proposed [6,7] and supported by experimental evidence that electrons assumed to be released with kinetic energy nearly zero can be driven by the field back to the vicinity of the core gaining a maximum of energy equal to $3.17U_P$. Such electrons can scatter elastically or inelastically, but since they are dressed by the field, one could invoke the Kroll and Watson theory [18,19]. In the Born approximation the important factor concerning the momentum transfer is $J_l^2(\Delta q a_0)$ where $\Delta q = q' - q$, q and q' are the initial and final momenta of the electron, respectively, $a_0 = \varepsilon/\omega^2$, and $(q')^2 - q^2 = 2l\omega$ for electrons which are emitted in the direction of the laser polarization in atomic units. For $q' \gg q$ we would have $\Delta q = q' - q \cong \sqrt{2l\omega}$. Finally l is the number of absorbed or emitted photons and J_l the Bessel function. The energy of the electron returning to the nucleus is at most $3.17U_P$ which at 8×10^{13} W/cm^2 is only a few electron volts. For $\varepsilon = 0.05$ a.u. and $\omega = 0.07$ a.u. the function $J_l^2(\sqrt{2l\omega}\varepsilon/\omega^2)$ has a significant maximum at $l = 10$, which coincides reasonably well with the simulation. At higher field strength, several maxima of J_l^2 appear up to $l \approx 8U_P/\omega$, a result that can be derived analytically. They would be washed out in an experiment because of the smooth pulse intensity shape, which is in accordance with the experimental data. The above argument is simply meant to make a qualitative connection, since it refers only to elastic scattering.

The picture would then be that some electrons are produced in a first step with zero kinetic energy. In a second step, they are driven by the electric field away and back to the core. There they may undergo a transition to the ground state and produce harmonics, or they may catch several additional photons and leave the atom. Accordingly, an ATI spectrum could now be interpreted as a superposition of the normal spectrum of electrons with those which absorbed additional photons. Whatever the details of the mechanism may be, it surely is not an accident that in Xe—where the recent lobes in the photoelectron angular distributions (PAD) have been observed [11]—our plateau appears at approximately the same photoelectron energy. In fact, we have already obtained data in Xe and Ar showing side lobes in PAD at the energy where the plateau begins. Details will be published elsewhere.

For hydrogen, everything is inherently included in the TD calculation, and the above argument is useful only in the sense of supporting a physical picture. For atoms other than hydrogen and especially the heavier rare gases, it is not obvious that the driven electron and the field do not affect the core by exciting and/or ionizing other electrons. It is in fact known [20,21] that even in He the second electron can be ejected by a process other than sequential multiphoton ionization, although at a frequency and intensity much different from ours. Thus a TD calculation based on a single active electron (in a frozen core) would not include inelastic scattering processes of the dressed electron. Part of such inelastic scattering would lead to double electron excitation which should affect the photoelectron spectrum. The energy range of the plateau is certainly within the manifold of doubly excited states in Xe, Kr, and Ar and their possible role in the overall behavior of the ATI spectrum is one of the intriguing questions raised by this work.

The possible role of ATI from the ions should finally be addressed. For this, the ionization of the atom must saturate before the peak of the pulse. Although for Kr and Xe this may have been the case, it should not have been a problem in He, Ne, and Ar [22]. In any case, one could imagine the superposition of photoelectron peaks from the atom and ion leading to an envelope exhibiting a change of slope. It cannot, however, exhibit a plateau unless the spectrum of either the atom or ion exhibits a plateau. Thus although photoelectrons from the ion may have contributed or even dominated the spectrum for high photoelectron energies, particularly in Xe, it cannot be the explanation of the basic effect. In other words, the effect would be present in the ion as well as in the atom.

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