

Atoms in Strong Optical Fields: Evolution from Multiphoton to Tunnel Ionization

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Electron energy spectra from ionization of the noble gases by 617-nm, 100-fs intense laser pulses show that the periodical double structure of narrow Stark-induced resonances and above-threshold ionization disappears gradually from xenon to helium. This implies that shifts and widths become of the order of the atomic-orbital frequencies, as expected at the onset of the tunneling regime.

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The behavior of atoms under strong irradiation has been a subject of constant interest ever since powerful laser sources became available. Although it is now relatively easy to produce extremely intense electromagnetic fields from compact, laboratory-sized laser systems, it is not possible to submit a given atom to an arbitrarily high intensity because it will be ionized before sensing the peak of the pulse. The maximum practical intensity, the saturation intensity, as defined by Lambropoulos [1], that can be applied to a neutral atom is determined by its ionization probability and the pulse characteristics. The upper limit for the saturation is reached by using state-of-the-art intense 70-fs pulses. The highest saturation intensities are obtained, under given laser conditions, with atoms having the highest ionization potential (e.g., helium). However, the ionization rate is too small to allow a study of the interaction at low intensities. A qualitative idea of the atomic behavior over a large range of intensities can nevertheless be obtained by studying similar atoms with increasing ionization potentials. The rare gases are well suited for this purpose and conveniently provide targets which span a factor of 2 in ionization potential and more than 10 in saturation intensities.

Ionization is one of the possible outcomes of the light-atom interaction. In the regime of strong fields, multiphoton ionization (MPI) and tunnel ionization (TI) are the two limiting cases of the ionization process [2]. The ratio γ of the tunneling time (i.e., the width of the barrier divided by the electron velocity) to the optical period is known as the adiabaticity or Keldysh parameter and is generally used to separate the two regimes,

$$\gamma = (IP/2U_p)^{1/2}, \quad (1)$$

where IP is the atom ionization potential and U_p the ponderomotive potential of the laser.

In the low frequency limit ($\gamma \ll 1$), TI is a good description of the transition dynamics. This is typical of rare gases ionized by a CO₂ laser, in which case the electron tunnels out in a time less than half the field period while its energy and momentum are subsequently deter-

mined by the Lorentz force [3]. In the other limit ($\gamma \gg 1$), the multiphoton character is evident in the photoelectron spectra as a structure repeated with the photon energy period [above-threshold ionization (ATI)] and whose rate scales as I^N , where I is the intensity and N the number of absorbed photons. This is typically the case for visible or uv excitation of an alkali atom. The ionization of rare gases by visible or near-infrared light spans a range of γ 's large enough to encompass both limits. According to the definition above, γ seems to be an increasing function of IP. However, γ is an intensity-dependent parameter through U_p . Therefore, the effective value of γ depends on the intensity at which the ionization is actually observed. It is qualitatively obvious that this intensity is an increasing function of the ionization potential. A quantitative estimate (known to be in good agreement with the experiment [4]) can be obtained by taking the threshold intensity for a (dc) field ionization,

$$I_{th} = IP^4/16Z^2, \quad (2)$$

in the definition of U_p in (1). The result of this substitution is that γ actually scales as $IP^{-3/2}$ and therefore decreases from xenon to helium. Furthermore, due to saturation, the effective γ in an experiment depends also on the pulse rise time. For short pulses, good agreement with the tunnel ionization rate [5] was found [6,7].

To study ionization, one possible option is to measure the power law of the total yield as a function of the incident intensity. However, it is also well known that such measurements are difficult and may lead to inconclusive results. The energy spectra of the photoelectrons resulting from the interaction between a laser pulse and an atom are known to carry somewhat more information than the ions. Many features that had remained hidden in total ion yield measurements have been uncovered by electron spectroscopy: the nonperturbative character of ATI, Stark-induced resonances (Freeman *et al.* [8]), etc.

These were the two basic motivations of the current work, in which we try to reconstruct a typical atomic response to strong fields over a large intensity range from

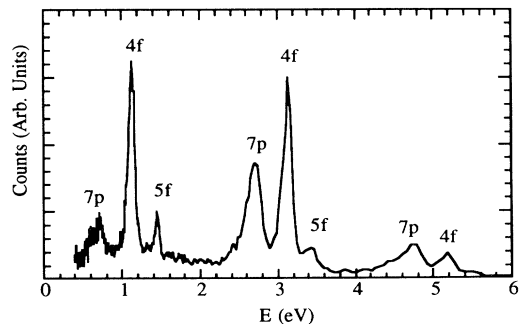


FIG. 1. Electron energy spectrum from xenon at $6.2 \times 10^{13} \text{ W cm}^{-2}$. Note that the resonances are reproduced with the photon energy period (2 eV).

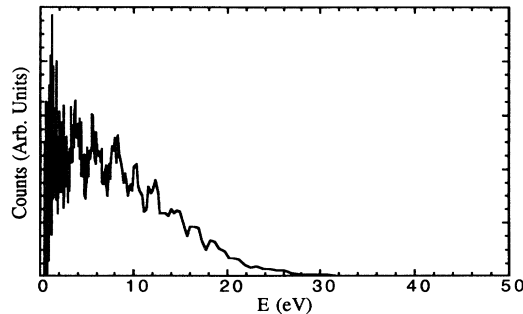


FIG. 2. Electron energy spectrum from neon at $6 \times 10^{14} \text{ W cm}^{-2}$. ATI is still seen superimposed on a large background.

the spectra obtained with the different rare gases. Briefly, a colliding pulse mode-locked oscillator is amplified in five Bethune dye cells and produces a maximum output energy of 2 mJ per pulse at a repetition rate of 20 Hz and a pulse duration as short as 75 fs at 617 nm. The linearly polarized beam is focused inside the interaction chamber by an achromatic doublet with an f number of 3, giving rise to intensities in excess of $5 \times 10^{16} \text{ W cm}^{-2}$. The electron spectrometer is a 1-m-long time-of-flight tube with a parabolic mirror electrostatic optic [9] which has a collection efficiency larger than 50% and a resolution of 50 meV at 1 eV. Electrons are detected by a dual 50-mm-diam multichannel plate detector with single-electron detection capability and signal processing is carried out by a digital oscilloscope with a 400-MHz sampling rate. The background pressure in the spectrometer is 2×10^{-9} torr. The target gas is leaked in the chamber at pressures varying from 10^{-7} to 10^{-4} torr which ensures a constant signal of a few counts per laser shot. The shot-to-shot pulse energy is monitored by a photodiode and this signal is used to store the electron data in up to ten different bins. Thus, it is possible to reduce the intensity fluctuations for a given bin to a few percent. The various bins are filled by varying the intensity with a half-wave-plate-polarizer combination and the natural shot-to-shot fluctuations.

By setting a negative voltage on the flight tube while keeping the parabolooids at ground, a field is created inside the parabola and ions can be collected and mass analyzed. The typical resolution is about 100 at $M=132$ (the xenon isotopes are barely resolved). This has been used to analyze the background ionization, to ensure that only one charge state is obtained (negligible multiple ionization), and to measure the total yield.

The evolution of the photoelectron energy spectra from xenon to helium is summarized in Figs. 1 to 3. The spectra of the lowest IP gases, xenon (IP=12.13 eV) (see Fig. 1), krypton (IP=13.99 eV), and argon (IP=15.76 eV), show a series of Stark-induced resonances which are reproduced in the different ATI orders. The resonances obviously dominate the transitions and, for xenon and ar-

gon, the spectra are relatively robust against intensity changes. As the peak intensity is increased beyond the appearance intensity, more resonances appear towards low energies but the spectra maintain their general structure. Therefore, from xenon to argon the ionization process keeps a clear multiphoton character in the sense that both the atomic structure and the photon energy play the major role and are clearly apparent on the spectra.

In xenon, the now well-known six-photon resonances of the nf states are observed ($n=4,8$). The appearance intensity is about $0.4 \times 10^{13} \text{ W cm}^{-2}$ corresponding to the $8f$ resonance. A small contribution from the p states is also detected. Another resonance appears at $3.9 \times 10^{13} \text{ W cm}^{-2}$ (Fig. 1) due to the $7p$ state. As the peak intensity is increased beyond $6 \times 10^{13} \text{ W cm}^{-2}$, the spectra show essentially no change in structure for a given ATI order but the relative amplitudes of the different orders are changing. The dynamics of the ATI structure in the presence of resonances will be analyzed elsewhere to keep this paper focused. The resonant processes are clearly dominant over the nonresonant ones, as seen from the low background between the resonant structure.

In krypton, the appearance intensity is $1.5 \times 10^{13} \text{ W cm}^{-2}$ and, again, the resonances dominate. However, the intensity behavior of the spectra is in strong contrast

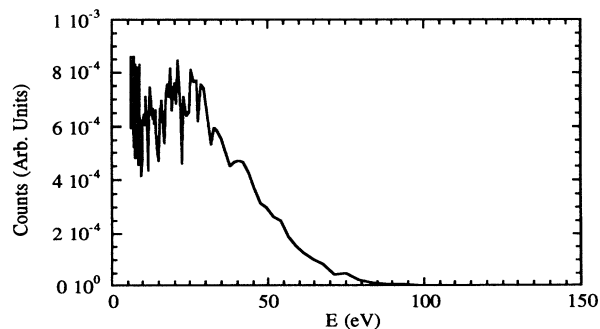


FIG. 3. Electron energy spectrum from helium at $1.5 \times 10^{15} \text{ W cm}^{-2}$. ATI and resonances have disappeared similar to the case of tunneling.

with the previous ones: The resonances merge into a continuum up to $9 \times 10^{13} \text{ W cm}^{-2}$ and a strong resonance, identified as the $4f$ state, eventually emerges for intensities beyond this value. This corresponds to an exceptionally large Stark shift, as discussed in detail by Mevel *et al.* [10] due to a combination of favorable factors (large static detuning of 2.9 eV) and high saturation intensity. In spite of this unusual behavior, the ionization process remains undoubtedly multiphoton.

In argon, the spectra again show outstanding resonances and behave similarly as the intensity is increased. Quantitatively, the intensity necessary to obtain spectra is $1 \times 10^{14} \text{ W cm}^{-2}$, but the appearance intensity is $2.5 \times 10^{13} \text{ W cm}^{-2}$. The saturation intensity is about $2 \times 10^{14} \text{ W cm}^{-2}$.

In contrast, the two lightest rare gases present spectra that differ drastically from the previous ones. Let us consider neon first. Its IP is significantly higher (21.56 eV) and so is the appearance intensity ($1.6 \times 10^{14} \text{ W cm}^{-2}$). The electron energy spectrum (Fig. 2) is observed up to an energy of 50 eV and presents a series of broad peaks whose separation is the photon energy. Some substructures still seem to be present in the spectra but they have not been identified. In conclusion, the multiphoton character is still obvious.

Finally, in helium, the appearance intensity is about 40 times larger than for xenon ($2.5 \times 10^{14} \text{ W cm}^{-2}$). As shown in Fig. 3 the electron spectrum, which extends to 90 eV, does not show structure above 30 eV. The chaotic spiking seen below is not reproducible, difficult to smooth out, and showing no relation with the photon energy. Checks have been carried out to ensure that structures are not washed out by space charge broadening. The most straightforward one is to leak in xenon gas at the same time as helium and record the spectrum at the helium intensity. The well-identified xenon structures remained quite resolved, thus demonstrating that their absence in the helium spectrum is not connected with an instrumental artifact. The contribution from impurities in the helium bottle (mostly H^+ , H_2^+ , H_2O^+) was also checked by recording mass spectra. They do not contribute more than a few percent to the total ion signal. Finally, broadening by ponderomotive acceleration is ruled out since the pulse turns off much faster than the time for electrons to escape the beam focus. For instance, at $1.5 \times 10^{15} \text{ W cm}^{-2}$, the drift velocity is $4.3 \times 10^6 \text{ m s}^{-1}$ and the electron exit time for a $10\text{-}\mu\text{m}$ focus is 1.15 ps.

This type of spectrum is strongly reminiscent of typical spectra in the long wavelength limit [3], that is, of field or tunnel ionization. This view is also supported by the measured ion yield which scales as I^5 , in good agreement with the prediction for tunneling [5]. In the low frequency limit, ionization is a quasi-instantaneous process leaving the free electron with a zero velocity. The electron final kinetic energy is determined by the classical motion in the electromagnetic field and the time at which the

electron is created [3]. In the case of linear polarization, this time is a fraction of the optical period around the crests and the troughs of the field oscillations. However, the disappearance of all structures in the spectrum is not, *per se*, a proof of the tunneling regime. Actually, a Floquet calculation in the case of hydrogen [11] shows that resonant structures may still be visible as the tunneling regime is approached. However, at intensities above the critical intensity (at which the saddle-point potential becomes equal to the electron binding energy), the ionization occurs in times of the order of $2\pi/\omega_{\text{at}}$, where ω_{at} is the atomic-orbital frequency. The Floquet calculation becomes inadequate then but still gives an indication of the widths. As they are much larger than the photon energy, it implies that all structures are washed out [11].

One question is whether or not the evolution of the spectra correlates to the variation of the adiabaticity parameter. Using the experimental intensities, one gets the γ values shown in Fig. 4. The upper and lower values correspond to the experimental appearance and saturation intensities, respectively. It is clear that there is some correlation since values for xenon to krypton are above unity, while for neon and helium they reach values below 1 where the tunneling regime begins. However, the γ values can be only indicative since they remain close to 1 contrary to the case of CO_2 laser experiments [3]. On the other hand, the electron spectra are significantly different.

In summary, this experimental study of rare gases ionization by intense 100-fs, 617-nm laser pulses by electron spectrometry shows the following trend: At the lowest intensity, the ionization process is unambiguously of multiphoton character and the Stark-induced resonances are dominant. States with Stark shifts as high as 2.9 eV are identified and the corresponding resonances are still outstanding. At an intensity about 10 times larger, the resonances are lost but the ATI structure is still apparent. Eventually, even this structure is lost as expected when

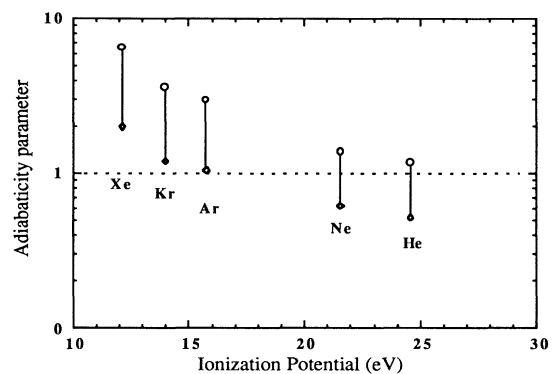


FIG. 4. γ values corresponding to appearance (○) and saturation (◇) intensities. The solid lines indicate the range of values accessible to experiment for each gas.

the shifts and widths become comparable to the atomic-orbital frequencies and at the onset of the tunneling regime, when the ionization process concentrates in times of the order of the optical period. Both the spectrum and the dependence of the rate as I^5 support the view that the tunneling regime has been reached. It is the first time, to our knowledge, that this transition has been observed at optical frequencies. It is a significant outcome of this paper that the results obtained in the infrared are confirmed in an experiment where the energy resolution is significantly smaller than the photon energy. This trend, from xenon to helium, may be viewed as the typical behavior of an atom submitted to high intensity radiation, if it could be recorded without the saturation intensity limitation.

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