Above-Threshold Ionization at the Few-Cycle Limit

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Photoelectron spectra measured for rare-gas atoms ionized with intense few-cycle laser pulses are presented. Several aspects of the few-cycle regime are discussed. In particular, the persistence of the plateaulike structure of spectra for high electron energies is shown. In contrast, a resonancelike feature at similar electron energies is suppressed as compared with longer laser pulses. Differences in the behavior of different species and implications for the electron-ion scattering cross section are pointed out.

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Investigation of atoms in strong laser fields has benefited enormously from the rapid progress in femtosecond laser technology in the past decade. One of the particularly remarkable achievements is the generation of pulses lasting two or fewer optical cycles (''few-cycle pulses''), i.e., full-width half-maximum pulse durations well below 10 fs at a center wavelength λ of 0.8 μm [1].

With respect to atomic behavior, use of sub-10 fs pulses makes a qualitative difference as compared with 30 fs pulses, because all Rydberg states have considerably longer Kepler periods than the pulse duration. As a consequence, resonantly enhanced multiphoton ionization, which plays a major role for longer pulses, is expected to be eliminated for few-cycle pulses. In this respect we note that for intense fields resonances are usually induced by light shifts of atomic levels which can easily exceed the photon energy. For a laser pulse with sub-10 fs FWHM duration the intensity varies from half the peak intensity to the peak intensity and back to its half in about three optical cycles. Accordingly, the atomic levels shift by energies bigger than the photon energy within one optical cycle or less, provided the concept of light shifts remains meaningful in the few-cycle regime. This means that the resonance condition can be fulfilled for hardly more than one optical cycle.

There is another advantage of extremely short pulses: State-of-the-art femtosecond lasers easily produce intensities exceeding 1 a.u. (3.5×10^{16} W/cm^2). Already at intensities 1 or more orders of magnitude lower, even rare-gas atoms ionize on the time scale of an optical cycle. Thus, the only way to expose atoms to high field strengths is to use the shortest pulses. This, in fact, has been one of the approaches to generate coherent soft-x-ray radiation reaching the water window by high-harmonic generation (HHG) [2]. However, due to the essential influence of macroscopic effects (phase matching), HHG is less favorable for investigation of fundamental questions of atoms in strong laser fields.

In this Letter, photoionization by intense 7 to 8 fs long laser pulses is considered. At intensities around 10^{14} W/cm^2, more photons than necessary for ionization can be absorbed by an atom, leading to photoelectrons with kinetic energies much higher than the photon energy (1.55 eV). This effect is called above-threshold ionization (ATI) [3]. We are particularly interested in the high-energy part of the ATI photoelectron spectra, which exhibits a characteristic plateaulike structure, i.e., an ionization probability independent of the photoelectron's energy up to a rather sudden cutoff [4]. The ATI plateau has been explained as being due to electrons returning to the ion core during the ionization process and rescattering elastically there. However, the role played by atomic resonances [5] and by the symmetry of the ground state [6] in this process is still under discussion.

Here, we show that the ATI plateau exists for few-cycle laser pulses and discuss the implications with respect to atomic resonances and other resonancelike effects investigated in a number of recent papers. Furthermore, evidence is given for different strengths of the plateau for different atoms. We attribute this to the cross section of ions for electron scattering.

It should be stressed that for few-cycle pulses the temporal variation of the laser's electric field depends on the phase φ of the carrier frequency with respect to the pulse envelope. Such phase effects have recently been detected [7]. However, most phase effects disappear very quickly as the pulse duration increases above 6 to 7 fs [8]. We carefully proved that phase effects do not play any appreciable role in the data presented, at least to the degree of sensitivity of the correlation scheme described in [7].

The experimental setup consists of a few-cycle femtosecond laser system and a combined photoelectron/ion spectrometer. The laser system has already been described and used in a number of previous experiments. In short, it consists of a 10-fs Ti:sapphire oscillator, a multipass

amplifier, and a prism compressor. The laser pulses thereafter are spectrally broadened in a gas-filled hollow fiber and compressed to 7 to 8 fs by means of several bounces on chirped mirrors. The pulse energy is about 400 μJ at a repetition rate of 1 kHz. With appropriate focusing mirrors a wide range of intensities can be realized. In order to adjust the intensity continuously in a precise way and without changing the pulse duration, we used a computer-controlled achromatic half-wave plate before the fiber and a Brewster plate (SF 10 glass) behind the fiber. Although most of the laser power is lost in this way, it is still sufficient to achieve up to 5×10^{14} W/cm^2 with $f =$ 250 mm focusing mirrors. Another method is to shift the laser focus with respect to the target gas. In this case, losses are negligible. The accuracy, however, is also lower because the beam geometry is not precisely known.

The spectrometer is based on the time-of-flight principle and permits simultaneous recording of photoelectrons and ions. The latter were used to record mass spectra and ensure that the photoelectron spectra are virtually free of contributions from any background gas. Two coaxial Mu-metal tubes in a UHV apparatus provide a field-free environment and form the flight distance (0.4 m) for the electrons. The target gas is fed to the center of the tube with a fine nozzle. Perpendicularly to the tube and gas stream the laser beam is focused on the gas cloud through appropriate small holes in the tube. Laser polarization is linear and parallel to the spectrometer axis. The electrons are detected at the end of one side of the tube and the ions at the other end by microchannel plates. In order to accelerate the ions, after each laser shot a high-voltage pulse is applied to a pair of metal plates with slits (width 250 μm) which are mounted symmetrically around the nozzle (slit-nozzle distance 10 mm) and perpendicularly to the tube. The HV pulse is delayed with respect to the laser pulse by 5 μs in order to ensure field-free conditions for the electrons. The time of flight is measured by two 2-GHz multiscalers (FAST 7886). The computer hosting the multiscalers also controls the half-wave plate. This allows quasisimultaneous recording of a series of intensities with precisely known ratio. The pinholes used to accelerate the ions also help to confine the laser intensity at which the recorded electrons were generated [9].

Figures 1 and 2 display ATI spectra for different rare-gas atoms at intensities between 0.5×10^{14} and 2.7×10^{14} W/cm^2. A first remarkable feature is the absence of virtually any ATI peak structure in the spectra. One might be tempted to attribute this to the shortness of the laser pulses in the sense that photoemission from a single cycle dominates. This, however, is not true in general, as has been shown in [8]. Rather, the spatiotemporal intensity distribution of a laser focus has to be considered: For locations in the focus, where the intensity is high, the ionization threshold experiences a considerable light shift and leads to a redshift of the photoelectrons generated there [11]. The light shift of the ionization

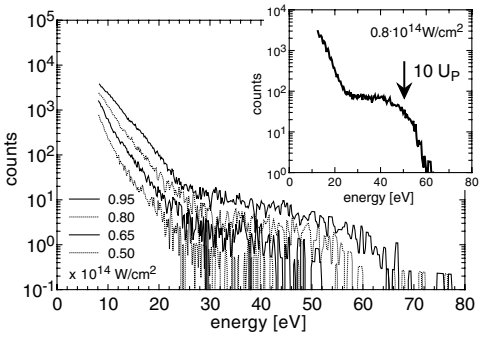

FIG. 1. Photoelectron spectra of xenon ionized with few-cycle laser pulses (≈ 8 fs) of different intensities. The intensity was calibrated with the spectrum shown in the inset and the $10U_P$-cutoff law.

threshold (and thus the redshift of the spectra) equals the ponderomotive energy U_P, which is defined as the quiver energy of a free electron in an oscillating field and proportional to the intensity and the wavelength squared. At 10^{14} W/cm^2, one has $U_P \approx 6$ eV, this being considerably higher than the photon energy and thus the spacing of the ATI peaks. A higher ionization yield in the center of the focus is compensated by the bigger volume of iso-intensity shells outside the center (''volume effect''). As a consequence, no ATI peak structure is expected for intensities around or higher than 10^{14} W/cm^2 and $\lambda =$ 800 nm.

This line of argumentation also holds for longer pulses, of course. The fact that ATI peaks are nevertheless observed there has to be traced back to particular regions in the laser focus, in which the intensity has a value such that the light shifts of atomic levels lead to particularly efficient ionization. One example is resonantly enhanced multiphoton ionization induced by the ponderomotive shift [12]. From the absence of any appreciable ATI peak structure in all of our measurements it can be concluded that atomic resonances do not play a role.

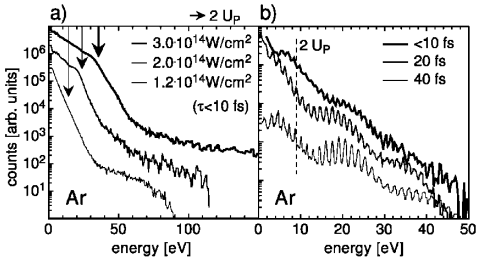

FIG. 2. (a) ATI spectra of argon at different intensities. Arrows indicate the energy position $2U_P$. (b) ATI spectra of argon for different pulse durations. The spectrum corresponding to 40 fs was taken from [10]. The clear enhancement of ATI peaks between 15 and 30 eV disappears as the pulse duration is decreased. The laser intensity for all spectra is approximately the same within 20% and equals 0.8×10^{14} W/cm^2. (The curves in both panels are separated in the vertical direction for visual convenience; the ratio of the curves thus has no meaning.)

Figures 1 and 2(a) also show that the ATI plateau exists in the few-cycle regime. In order to test the hypothesis that the observed plateaus for "long" and few-cycle pulses are due to the same mechanism—namely, rescattering of electrons returning to the ion core within one optical cycle after initial ionization—measurements with circularly polarized few-cycle laser pulses were performed. The additional component of the electric field would prevent electrons from returning to the ion core.

Circular polarization was generated by introducing an achromatic quarter-wave plate (Halle, Berlin) before the hollow fiber. There, the bandwidth of the wave plate is big enough to create circularly polarized light of high quality. The disadvantage of the approach is that the pressure in the fiber has to be changed as the ellipticity is changed in order to preserve few-cycle pulses. Therefore, systematic measurements at other ellipticities are cumbersome. The result of our measurements with circular polarization was that the ATI plateau indeed disappears at circular polarization; see Fig. 3.

The observation that the ATI plateau has a similar behavior for long and for few-cycle pulses leads to the conclusion that the ATI plateau is not a resonance phenomenon as proposed in [5]. Figure 1 displays the dependence of Xe-ATI spectra on laser intensity. Similar to experiments with longer pulses, the ATI plateau extends to higher energies for higher intensities. The energy of the end of the plateau (cutoff energy) can be calculated to lie at $10U_P$ for long pulses [13]. This gives a very reliable handle to calibrate the intensity. For few-cycle pulses, the ATI cutoff energy in principle depends on the phase φ. However, this effect is smaller than 10% for pulses longer than 7 fs. Therefore, the ATI cutoff again can serve to determine the applied peak intensity with an estimated error of about 10% as long as the plateau structure is clear enough. Intensity estimations by conventional means give compatible results, however, with considerably bigger uncertainties.

Additional evidence for the applicability of classical considerations can be found at lower electron energies. Figure 2(a) displays argon spectra recorded at intensities between 1.2×10^{14} and 2.7×10^{14} W/cm^2. As the intensity is increased, the low-energy part of the spectra develops an increasingly clear kink at electron energies corresponding to $2U_P$. This classical limit for electrons leaving the atom without rescattering (so-called "direct" electrons) can easily be derived by assuming that an electron is lifted into the continuum at some phase of the laser's electric field and starts from the atom with zero velocity. Electrons starting at phases where the field crosses zero will acquire a drift energy of $2U_P$ [14]. So far, clearer evidence for the validity of this law has been found only in microwave ionization experiments [15]. For these, $U_P/(\hbar\omega)$ is many orders of magnitude larger and classical behavior is expected.

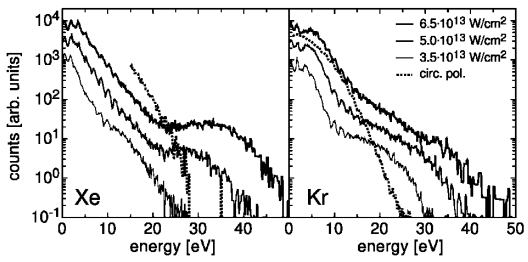

FIG. 3. Comparison between xenon (left panel) and krypton (right panel). The relative height of different spectra is not of significance. For comparison, spectra for circular polarization (dotted curves) are shown. They correspond to intensities of $\approx 9 \times 10^{13}$ W/cm^2 (Xe) and 7×10^{13} W/cm^2 (Kr).

For intensities considerably higher than 10^{14} W/cm^2, one observes that the plateau degrades to a change in slope [Fig. 2(a), 2×10^{14} W/cm^2 and 3×10^{14} W/cm^2]. This effect is again similar to the situation for long pulses. For an explanation, the spatiotemporal profile of the laser focus has to be considered once more: The lower the electron energy, the more focal volume can produce such electrons and thus the stronger the yield for these energies.

Recently, resonancelike behavior in the yield of plateau electrons [4,16,17] has attracted a lot of attention. At certain intensities, the low-energy part of the plateau changes by up to an order of magnitude if the intensity is varied by less than 5%. Experimentally, the effect was found to be particularly strong for argon. All spectra recorded for intensities higher than the resonance intensity exhibit the feature, because the resonance intensity is then realized somewhere in the laser focus. Therefore, for a sufficiently strong resonance, the phenomenon is easily detected experimentally. Two different explanations have been found. The first one invokes multiphoton resonances with atomic levels which are shifted into resonance due to the ac-Stark effect [5,18]. An alternative explanation traces the resonances back to channel closings [10,19], i.e., to intensities where the continuum limit is shifted into resonance. It appears likely that both possibilities are realized in nature: atomic resonances for long pulses and channel closings for pulses shorter than about 35 fs. For the latter, very few optical cycles are within that region around the pulse maximum, for which the field amplitude changes by not more than a few percent. Accordingly, the levels of an atom in a short laser pulse are so broad that the atomic Rydberg levels merge. This creates a sudden change in the density of states below the ionization threshold, which have been shown to cause resonancelike effects in the ATI plateau.

Interestingly, both alternatives as an explanation arrive at the conclusion that constructive interference of wave packets oscillating for many optical cycles in the vicinity of the atom are the actual reason for the resonancelike behavior. The controversy is just the question what causes very different wave packet orbits to interfere constructively. Using few-cycle pulses, it is possible to verify the common aspect of the two theories. Few-cycle pulses are

so short that no orbits longer than about two laser cycles can play any appreciable role. The expectation therefore is that the resonancelike effect does not exist for few-cycle laser pulses. This is indeed the case. By simply evacuating the hollow fiber and thus generating 20-fs pulse the resonances reappeared, although clearly weaker than for the 40-fs pulses used in previous experiments [10]; see Fig. 2(b). This is in agreement with the prediction of [18].

As mentioned above, the resonancelike effect is manifested very differently for the various rare-gas species. So far, theory and experiments for strong-field laser atom experiments have hardly addressed the question of differences between the atomic species. Rather, most of the approximations used emphasize the common aspects by neglecting atomic structure. This holds, in particular, for the strong-field approximation in its classical and quantum version. Evidence for qualitatively different behavior of neon and argon with respect to nonsequential double ionization has been observed in recent momentum spectroscopy experiments [20].

Here we would like to draw attention to krypton. In Fig. 3, we show a comparison between krypton and xenon for different laser intensities. Spectra corresponding to different species but the same intensity were recorded by switching between the gases as quickly as possible and without changing anything else. The time needed to switch between the gases was of the same order as the time needed to record the spectra, i.e., several minutes. No severe drifts in the performance of the laser were observed within these short periods of time. In contrast to the other rare gases, krypton spectra do not exhibit clear ATI plateaus for intensities above 5×10^{13} W/cm^2. Only for lower intensity can short plateaus be observed [21]. This peculiarity can also be seen for 50 fs laser pulses and at a wavelength of 630 nm [4]. However, it has never been possible to provide more than speculation for the underlying mechanism. Since processes involving atomic structure cannot play a role for few-cycle pulses and because they should change when switching from long to few-cycle pulses and from $\lambda = 630$ nm to 800 nm, they are very unlikely to be responsible for the absence of pronounced plateaus in krypton ATI spectra. Rather, the reason should be sought in the scattering cross section of the krypton ion. On the basis of the ATI spectra measured, we predict that the elastic scattering cross section of krypton ions is small for electron energies above 30 eV. A similar conclusion has been drawn from results of experiments with alkali atoms exposed to mid-infrared radiation [22].

It should be noted that ATI plateau electrons return to the ion core close to a zero crossing of the electric field. Therefore, the electron scatters in nearly field-free conditions. Future experiments with phase-stabilized few-cycle laser pulses will allow one to tailor the conditions under which the electrons scatter.

In conclusion, we have investigated multiphoton ionization for intense few-cycle laser pulses. The presence of the ATI plateau confirms the rescattering scenario. The absence of resonancelike enhancement in the ATI plateau supports theories invoking constructive interference effects of long trajectories as an explanation. The absence of a pronounced ATI plateau in krypton is obviously due to a low scattering cross section of krypton ions for fast electrons.

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