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Above-threshold ionisation of He at 248 nm: the role of the ionic contribution

C.J.G.J. Uiterwaal[1], **D. Xenakis**[1], **D. Charalambidis**[1,2]

[1] Foundation for Research and Technology-Hellas, Institute of Electronic Structure & Laser, Laser and Applications Division, P.O. Box 1527, GR-71110 Heraklion, Crete, Greece (Tel. + 30-81/391125, Fax: + 30-81/391318, e-mail: kees@iesl.forth.gr)
[2] Department of Physics, University of Crete, Crete, Greece

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Abstract. Photoelectron spectra of rare gas atoms interacting with 0.5 ps KrF laser pulses at intensities up to 10^{16} W/cm^2 are reported. For intensities higher than the saturation intensity of the atom the envelope of the logarithm of the spectrum exhibits two different slopes with strong evidence that these originate from the superposition of the atomic and ionic above-threshold ionisation (ATI) spectra. At high intensities the ionic part of the spectrum becomes so flat that it appears plateau-like.

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1 Introduction

Interaction of an atomic system with strong laser pulses of short pulse duration results in photoelectron spectra that consist of a sequence of peaks separated by the photon energy [1]. The phenomenon, known as above-threshold ionisation (ATI), corresponds to the absorption of an integer number of photons above the ionisation threshold before the atom decays to an ion and a free electron. The peak heights in the sequence have been known for some years now to decrease for increasing photoelectron energies [2], at least as long as perturbation theory is valid. The envelope of the logarithm of the spectrum is then effectively a straight line, the slope of which becomes less steep for increasing intensities.

In a recent letter [3], a departure from this behaviour has been demonstrated. Ionisation of rare gases with strong short pulses was found to result in ATI spectra in which the slope of the envelope exhibits strong changes at different kinetic energy ranges of the spectrum, showing a plateau in some cases. A second unexpected observation in ATI spectra was the appearance of side lobes in the photoelectron angular distributions (PAD) for certain peaks [4, 5] where the AD was expected to be strongly peaked. A connection between the position of the plateau and the appearance of side lobes has also been established recently [5]. A model [6, 7] utilising electrons released with near zero kinetic energy that are first accelerated away and then driven back to the vicinity of the core by the field has been employed for the interpretation of the observations. Such electrons are considered to return back to the core with a maximum kinetic energy of $3.17\,U_p$, U_p being the ponderomotive shift. In the vicinity of the core they scatter while absorbing a number of additional photons thus giving rise to ATI [3]. The role of ATI from *ions* has also been considered [3] for the appearance of the plateau and has been concluded not to be the explanation of the basic underlying effect.

The purpose of this work is to present results of an experiment performed in He under different experimental conditions to the above, namely shorter wavelength and longer pulse duration, that show similar features of a however different origin. Briefly, the present experiment demonstrates the appearance of a second, less steep slope in the envelope of the ATI spectrum in the high kinetic energy region of the spectra, with strong evidence that this second slope is due to ATI of the ion of the parent atom. The evidence is provided from a series of different observations compatible with each other. The present work (*i*) indicates an increased importance of the role of ionic ATI in accounting for the detailed features of the photoelectron spectra, (*ii*) raises the question of the relative weights between the contribution of ionic ATI and that of other effects, including non-sequential double ionisation, to an observed plateau, and (*iii*) suggests that filtering out the ionic contributions may reveal the fingerprints of any other effects present in the spectrum, thus leading to more detailed information towards the determination of any other mechanism possibly responsible for the appearance of an ATI plateau.

2 Experimental set-up

The experiment is performed by utilising: (*i*) a hybrid dye-excimer laser system [8] delivering pulses of ~ 500 fs duration at a wavelength of ~ 248 nm, with a repetition rate of 4 Hz, a maximum energy of 14 mJ and an amplified

spontaneous emission (ASE) to pulse contrast ratio $< 10^{-6}$ at focus, and (*ii*) a magnetically shielded 20 cm long photoelectron time-of-flight spectrometer with an energy resolution $\Delta E/E \approx 7\%$ for photoelectrons with a few eV of kinetic energy. The signal of the microchannel-plates detector was recorded with a digital oscilloscope. The overall resolution of the system drops below the 5 eV photon energy for photoelectron energies above 60 eV, thus not allowing the ATI peaks to be resolved in this energy regime.

The background pressure in the spectrometer was 10^{-9} mbar. He gas with a purity of 99.9999% was employed. After passing through a liquid nitrogen trap, the gas was inserted into the vacuum chamber of the spectrometer by means of a continuous flow through a gas dosing valve. A quadrupole rest gas analyser was employed to monitor the composition of the gas in the vacuum chamber. The target gas pressure was varied from 10^{-7} to 10^{-5} mbar for runs at different laser power densities. The laser beam was focused into the target gas to laser power densities up to $\sim 10^{16}$ W/cm^2. Linear and circular polarisations of the beam have been used for comparison of the spectra.

3 Results and discussion

Figure 1 shows ATI spectra of He measured at different laser power densities. Each spectrum is an average of 1000 laser shots from which the background spectrum measured for each laser intensity has been subtracted. Care was taken to maintain optimal pulse-to-pulse stability of the laser intensity. At lower laser power densities the spectra consist of a series of peaks separated by the photon energy of about 5 eV (Fig. 1, left panel). No clearly resolved structure due to dynamical resonances [9] is observed, indicating that the ionisation cross section does not change abruptly during the temporal evolution of the laser pulse. For a certain range of laser power densities the ATI peaks are shifting linearly with the laser energy as expected due to the increasing ponderomotive shift of the ionisation threshold. An asymmetric broadening with a longer tail on the high energy side, compatible with the ponderomotive shift, can also be seen in the spectra. At about 10^{15} W/cm^2 the shift saturates to a constant value. This stabilisation of the shift can be attributed to the saturation of ionisation caused by a fraction of the leading edge of the laser pulse and can be used for the determination of the saturation intensity and an "*effective*" generalised multiphoton ionisation cross section as described elsewhere [10]. At higher power densities the peaks are becoming broader and strongly overlapping due to the shifts caused in the expanding interaction volume. Furthermore a photoelectron energy distribution with unresolved structure appears in the high kinetic energy part of the spectrum exhibiting an unambiguously smaller slope than the envelope of the ATI peaks (Fig. 1, right panel). At the highest measured laser power densities this distribution extends up to a few hundred eV, with such a small slope that it appears as a 'plateau' in the total photoelectron distribution. The proportion of the signal corresponding to the 'plateau' part of the curve increases with laser intensity. The two distributions intercept each other between 20 and 35 eV, depending on the laser intensity. The lack of clearly resolved structure in the high kinetic energy region, where the 'plateau' appears, can be partially attributed to the resolution of the detection systems at very high kinetic energies, as well as to the ponderomotive shift U_p which at such high intensities I is several times the photon energy and thus may smear out the ATI structure. For the present wavelength U_p is given by the relation $U_p = 5.7 \cdot 10^{-15} I$ (U_p in eV and I in W/cm^2).

Similar results have been obtained in other rare gases like Ar and Kr. These systems exhibit lower ionisation threshold and thus lower saturation intensity. These results will be presented in a forthcoming extended publication.

Figure 2 shows the dependence of the slopes of the two photoelectron distributions (envelopes) as a function of the laser power density. The slope of the envelope of the atomic ATI peaks is initially a linear function of the logarithm of the power density but becomes constant at higher intensities. The linear dependence would be compatible with lowest order perturbation theory, assuming that the ratio of the cross sections for two subsequent ATI peaks remains constant for all the peaks, although at the intensities employed one could expect a deviation from such perturbative behaviour. The stabilisation of the slope to a constant value can be attributed to the saturation of ionisation and is indeed observed for laser intensities above the saturation intensity of $9.5 \cdot 10^{14}$ W/cm^2 [10].

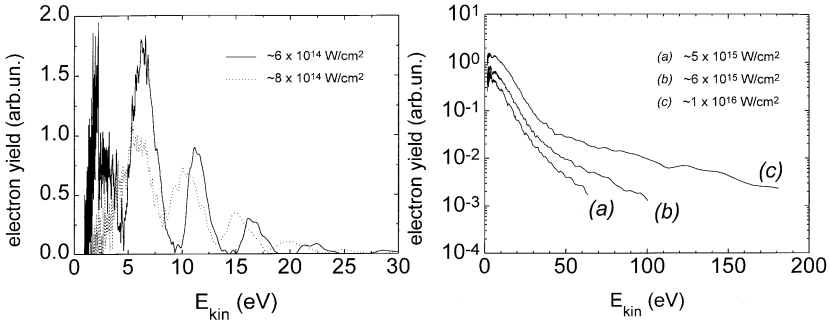

Fig. 1. ATI spectra of He recorded at different laser power densities. Note the differences in the scales of the *left* and *right panel*

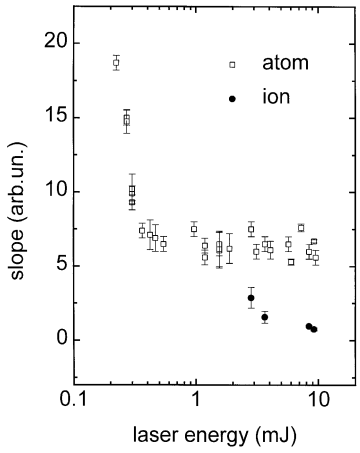

Fig. 2. The laser power density dependence of the two slopes of the photoelectron spectrum envelope. The saturation of the atomic ionisation is clearly depicted in the stabilisation of the corresponding slope value

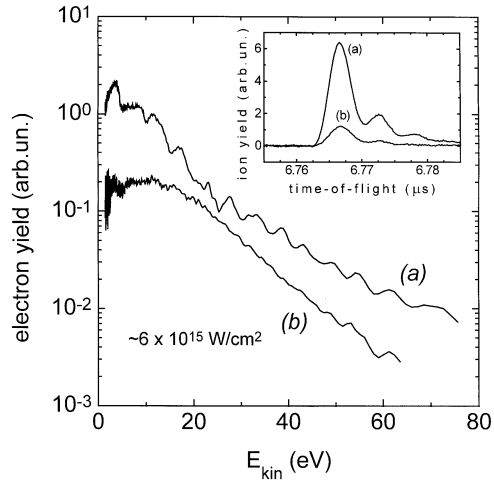

Fig. 3. Photoelectron spectra of He recorded with linearly (*a*) and circularly (*b*) polarised light at $\sim 6 \cdot 10^{15}$ W/cm^2. (The insert shows the corresponding difference in the total He^{2+} ion yield with linearly (*a*) and circularly (*b*) polarised light at $\sim 10^{16}$ W/cm^2)

The slope of the second distribution at high kinetic energies is also decreasing with laser power density.

The fact that the less steep slope is observed for laser intensities well above the ionisation saturation of the atom (five times higher) provides strong evidence that the ionisation of the H-like helium ion dominates this part of the spectrum. The observed distribution can be attributed to the ATI spectrum of He^+ in a sequential double ionisation process of He. An ionic contribution in ATI spectra of He has been previously considered for 780 nm, 100 fs pulses for intensities beyond $3 \cdot 10^{15}$ W/cm^2 [11]. Measurements of yields of singly and doubly ionised species versus laser power density at the wavelength of the present experiment [10] support the assumption of a sequential process that creates singly ionised species in the first step and subsequently removes a second electron in the second step. From these measurements we estimate the ratio of the total double to single ionisation yield to be of the order of 10% for the highest laser power densities employed in the present experiment. This is a significant amount of double ionisation that supports the argumentation of the present work. Far above the saturation intensity only ions are available, no atoms exist anymore in the interaction volume. Since He has a higher ionisation threshold and hence saturation intensity than any other element, any rest gas in the interaction volume has also reached saturation of ionisation. He ions are at least 10^3 times more abundant than any other atomic or molecular ion and are thus the only significant source for the new features (less steep slope in the high kinetic energy part of the spectrum) that appear only at high laser intensities. The different slopes for the atomic and ionic ATI envelopes can be attributed to the considerable difference in saturation intensity between the two species. The shape of the ionic curve in Fig. 2 could be an indication of departure from perturbative behaviour at these high intensities.

Measurements with circularly polarised light do not give any evidence for a second slope (Fig. 3b). In the spectrum of Fig. 3a the first four "oscillations" can be attributed to ATI peaks while the rest is convolution of ATI peaks with noise "oscillations". Remaining ATI structure can also be recognised in the low energy region of the spectra in the right panel of Fig. 1. Fine differences in the spectra of the two figures may arise because the spectrum of Fig. 3a was recorded using purified linear polarisation, in contrast to the spectra of Fig. 1, where no polarising elements have been employed in order to achieve the highest possible intensity. The results shown in Fig. 3 are similar to those of the experiment performed at 630 nm with 40 fs pulses [3]. They are also compatible with the presence of the ionic ATI in the spectra shown in Fig. 1. Because at least eleven photons are needed for the ionisation of He^+, contribution from very high total angular momentum states of the ion is required. Although not in every case true, this is in general expected to result in a reduced ionisation rate thus pushing the number of the ejected electrons below the detection limit of the experimental set-up. Further verification for this is provided by measurements of the total He^{2+} ion yield that show a strong signal reduction when the polarisation is changed from linear to circular at an intensity of $\sim 10^{16}$ W/cm^2 (see insert in Fig. 3).

In all investigated cases (except with circular polarisation, Fig. 3b), the ionic contribution leads to a distinct change of the slope of the envelope of the ATI peaks, dividing the spectrum in two parts. The slope never really becomes zero or positive as to form a clear plateau or a maximum in the envelope as is the case in [3].

4 Conclusions

The results of the present work strongly suggest an important contribution from the ionisation of the ion to the ATI spectrum of an atom exposed to a strong electromagnetic pulse. The contribution of ions in atom-strong field interactions has been considered in high-order harmonic

generation and is found to be important at short laser wavelengths [15]. A direct experimental proof of this contribution and quantitative comparison with single-atom calculations in higher-order harmonic generation is a rather complex issue due to phase variation and propagation effects. The present results in ATI introduce a more straightforward procedure for the investigation of the single-atom, single-ion response in high intensity interactions. The contribution of different ionic states should be considered as of major importance for all ATI spectra that exhibit deviations from the behaviour expected from lowest order perturbation theory, in particular in the high laser frequency regime. It should be pointed out that at 248 nm double ionisation is a sequential multiphoton processes [10, 12], while at around 600 nm it may become non-sequential as has been manifested in He [13, 14]. A further evidence of the different ionisation mechanisms taking place in the two different wavelength regimes is that in the present work no relation between the photoelectron energy at which the change of the slope occurs and a multiple of the ponderomotive shift is found in contrast to the case ($2U_p$) of the tunnelling non-sequential double ionisation of He at 780 nm [11]. Differences in detailed features of the results of the present experiment and those of the experiment at 630 nm [3] could speculatively be associated with this matter. The weight factor of the ionic ATI contribution to a plateau could be determined through more sophisticated experiments utilising ion-electron coincidence measurements for the different charge states produced during ionisation. In such an experiment the ATI spectrum of every charge state can be separated from that of all the other charge states, thus unravelling the different processes that contribute to deviations from lowest order perturbation theory in ATI spectra at high intensities.

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References

1. Agostini, P., Fabre, F., Mainfray, G., Petite, G., Rahman, N.K.: Phys. Rev. Lett. **42**, 1127 (1979)
2. Mohideen, U., Sher, M.H., Tom, W.K., Aumiller, G.D., Wood, II O.R., Freeman, R.R., Bokor, J., Bucksbaum, P.H.: Phys. Rev. Lett. **71**, 509 (1993)
3. Paulus, G.G., Nicklich, W., Xu Huale, Lambropoulos, P., Walther, H.: Phys. Rev. Lett. **72**, 2851 (1994)
4. Yang, B., Schafer, K.J., Walker, B., Kulander, K.C., Agostini, P., DiMauro, L.F.: Phys. Rev. Lett. **71**, 3770 (1993)
5. Paulus, G.G., Nicklich, W., Walther, H.: Europhys. Lett. **27**, 267 (1994)
6. Corkum, P.B.: Phys. Rev. Lett. **71**, 1994 (1993)
7. Schafer, K.J., Yang, B., DiMauro, L.F., Kulander, K.C.: Phys. Rev. Lett. **70**, 1599 (1993)
8. Szatmari, S., Schäfer, F.P.: Opt. Commun. **68**, 196 (1988)
9. Freeman, R.R., Bucksbaum, P.H., Milchberg, M., Darack, S., Schumacher, D., Geusic, M.E.: Phys. Rev. Lett. **59**, 1092 (1987); Rottke, H., Wolff, B., Brickwedde, M., Feldmann, D., Welge, K.H.: Phys. Rev. Lett. **64**, 404 (1990); Rudolph, H., Tang, X., Bachau, H., Lambropoulos, P., Cormier, E.: Phys. Rev. Lett. **66**, 3241 (1991)
10. Charalambidis, D., Xenakis, D., Uiterwaal, C.J.G.J., Maragakis, P., Jian Zhang, Schröder, H., Faucher, O., Lambropoulos, P.: (to be published)
11. Walker, B., Sheehy, B., DiMauro, L.F., Agostini, P., Schafer, K.J., Kulander, K.C.: Phys. Rev. Lett. **73** 1227 (1994)
12. Kondo, K., Sagiasaka, A., Tamida, T., Nabekawa, Y., Watanabe, S.: Phys. Rev. A **48**, R2531 (1993)
13. Fittinghoff, D.N., Bolton, P.R., Chang, B., Kulander, K.C.: Phys. Rev. Lett. **69**, 2642 (1992)
14. Walker, B., Mevel, E., Yang, B., Breger, P., Chambaret, J.P., Antonetti, A., DiMauro, L.F., Agostini, P.: Phys. Rev. A **48**, R894 (1993)
15. Preston, S.G., Sanpera, A., Zepf, M., Blyth, W.J., Smith, C.G., Wark, J.S., Key, M.H., Burnett, K., Nakai, M., Neely, D., Offenberger, A.A.: Phys. Rev. A **53**, R31 (1996); Kondo, K., Tamida, T., Nabekawa, Y., Watanabe, M., Watanabe, S.: Phys. Rev. A **49**, 3881 (1994); Krause, J.L., Schafer, K.J., Kulander, K.C.: Phys. Rev. Lett. **68**, 3535 (1992); Xu, H., Tang, X., Lambropoulos, P.: Phys. Rev. A **46**, R2225 (1992)