Experimental and theoretical study of 3-photon ionization $He(1s2s\,^3S)$ and $He(1s2p\,^3P)$

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While the photoionization of atoms in the ground state has been intensively studied, less is known concerning photoionization of excited states, particularly multiphoton ionization [1,2,3]. We report here the results of an experimental and a theoretical study of the three-photon ionization of the $1s2s\,^3S^e$ and $1s2p\,^3P^o$ states of helium by a laser operating in the green and red regions of the visible spectrum.

In the experiment, He^- ions are first formed in the $1s2s2p\,^4P^o$ state by collisions of fast helium ions with cesium. A pulsed dye laser pumped by the second or third harmonics of a Nd:YAG laser is then used to photodetach an electron, leaving an atom of helium in either the $1s2p\,^3P^o$ or the $1s2s\,^3S^e$ state. These are subsequently ionized by the absorption of three more photons. By tuning the wavelength of the laser, the ion yield from either of the two excited states can be measured. In the work reported here, the wavelength is varied within the 530-560 nm and 685-730 nm ranges in order to probe, respectively, the $1s2s\,^3S^e$ and $1s2p\,^3P^o$ states. Dependence of the ionization on the photon flux and the laser polarization is also studied by varying the laser peak intensity between $1.25 \times 10^9\,\mathrm{W\,cm^{-2}}$ and $3.6 \times 10^{10}\,\mathrm{W\,cm^{-2}}$ and by changing the polarization state from linear to circular. The experimental results show two series of asymmetric peaks, associated to two-photon resonances with ns and nd Rydberg states for $He(1s2s\,^3S^e)$ and with np and nf Rydberg states for $He(1s2p\,^3P^o)$. For the latter, a series of peaks has tails towards higher photon energies while the other has tails changing direction below 706.7 nm.

A model Hamiltonian [4] is built using matrix elements from DVR and QDT calculations and checked against a full, *ab initio* R-matrix Floquet calculation. The time-dependent Schrödinger equation is numerically integrated to reproduce the experimental spectra with different pulse peak intensities. The series of peaks are consistently reproduced over the large wavelength ranges considered, both in shape and position.

For the $1s2p\,^3P^o$ ($M_L = 0$) state, the $1s2p\,^3P^o$ and $1s3s\,^3S^e$ states are coupled by a one-photon interaction, and hence are strongly mixed over a relatively wide range of laser wavelengths. This dressing is absent in the case $M_L = \pm 1$. Ionization is shown to occur *via* two resonantly enhanced multiphoton ionization (REMPI) schemes: a (1+1+1) scheme for $M_L = 0$, never encountered before in atomic multiphoton ionization, and a (2+1) scheme for $M_L = \pm 1$. $He(1s2s^3S^e)$ exhibits a much simpler (2+1) REMPI behaviour as a function of the laser wavelength and intensity. The laser polarization has also an important effect on the ionization yield and highlights, as for the ionization of $He(1s2p\,^3P^o)$, the influence of the magnetic quantum number.

References

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