Experimental and theoretical study of 3-photon ionization of the 1s2p ^{3}P^o state of helium

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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 [1–3]. We report here the results of an experimental and a theoretical study of the three-photon ionization of the 1s2p ^{3}P^o state of helium by a laser operating in the red end of the visible spectrum.

In the experiment, He^- ions are first formed in the 1s2s2p ^{4}P^o state by collisions of fast helium ions with cesium. A pulsed dye laser pumped by the second harmonics of a Nd:YAG laser is then used to photodetach an electron, leaving an atom of helium in either the 1s2s ^{3}S^e or the 1s2p ^{3}P^o 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 from 685 to 720 nm in order to probe the 1s2p ^{3}P^o state, for laser peak intensity between 1.25×10^9 W cm^{-2} and 3.6×10^{10} W cm^{-2}. The experimental results below 705 nm show two series of two-photon resonances, one of which is blue-shifted as the intensity of the laser increases while the other is red-shifted.

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. For $M_L = 0$, the 1s2p ^{3}P^o and 1s3s ^{3}S^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 $M_L = 1$. The two series of two-photon Rydberg resonances are thus associated with the two initial states 1s2p ^{3}P^o($M_L = 0, 1$), and we conclude that their differing behaviour as a function of peak laser intensity is due to the strong dressing effects present when $M_L = 0$. We predict that for laser wavelengths above the one-photon crossing between the 1s2p ^{3}P^o and 1s3s ^{3}S^e states at about 706 nm, both resonance series will be blue-shifted with increasing laser peak intensity. Experimental verification of this behaviour is currently under way.

References:

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