Multiphoton processes involving laser-driven inner-shell electrons

H. W. van der Hart and M. Madine

Department of Applied Mathematics and Theoretical Physics, Queen's University Belfast, Belfast, BT7 1NN, UK

Main contact email address h.vanderhart@qub.ac.uk

Introduction

Over the last decades, great advances have been made in the experimental and theoretical investigation of the response of outer electrons to intense laser fields. Recent developments in laser technology include the development of intense lasers operating at 2 micron [1], and the development of X-ray lasers [2]. The techniques developed for 780 nm are suitable, at least in principle, for application to intense-field processes at 2 micron. However, they are not well suited for application to intense-field processes in the X-ray regime.

In the X-ray regime, the assumption that only the outer electron interacts with the laser field is no longer appropriate, since inner electrons can also interact with the laser field. For ground-state Li, for example, the removal of the outer 2s electron requires 5 eV, while the removal of the 1s electron requires about 60 eV. Hence, at a wavelength of 40 nm, only two photons need to be absorbed to remove the 1s electron. In order to describe the full response of an atom to an intense 40-nm laser field, the response of both the 2s electron and the 1s electrons must thus be taken into account.

In the present report, we discuss recent progress made in the description of the response of inner electrons to intense VUV or X-ray laser fields. The R-matrix Floquet approach was developed at Queen's University Belfast as a technique particularly aimed at describing the interaction between multi-electron atoms and intense laser fields [3]. Since the description of the response of inner electrons requires an approach capable of dealing with complex atomic structure, this approach would be among those best suited for this investigation.

We illustrate how R-matrix-Floquet theory can be used to describe inner-shell processes, first by studying multiphoton ionization of He in the excited 1s2s ^{1}S state [4,5]. Although this initial state is not commonly used in experiment, it allows an easier comparison of the response of inner-shell electrons versus the response of outer-shell electrons. We then investigate two-photon detachment of 1s and 2s electrons from the $1s^22s^2$ ^{1}S ground state of Li^-, a real multi-electron atom [6].

Comparison of the response of inner and outer electrons

The R-matrix Floquet approach combines standard R-matrix theory for electron scattering and photoionization [7] with the Floquet-Fourier Ansatz. The effect of this Ansatz is that the wavefunction is partitioned into so-called Floquet blocks, with each Floquet block describing the wavefunction after absorption of a certain net number of photons. All the atomic structure effects are contained within a Floquet block, while the laser field couples the Floquet blocks. The Floquet expansion typically contains a few blocks outside the processes of interest to ensure to convergence. In addition, the presence of the laser field destroys the radial symmetry of the atom, and hence each Floquet block contains an expansion over angular momentum.

The amount of atomic structure included is indicated by the so-called target states. For He, first calculations are carried out using a three-target-state basis, the 1s, 2s and 2p orbitals for He^+. This basis means that at least one of the He electrons must be in one of these 3 orbitals. To estimate the influence of finals-state resonances, we extend the target-state basis by adding the 3s, 3p and 3d orbitals of He^+. This basis allows the description of resonances such as $3s^2$ ^{1}S, which cannot be described using the more restrictive basis. For Li^-, we use a fourteen-target-state basis as described in [8].

Comparison of the response of inner and outer electrons

To compare and contrast the response of inner and outer electrons, we investigate the 1s2s ^{1}S state of He exposed to an intense laser field. Obviously, less energy is required to remove the 2s electron than to remove the 1s electron. In the present photon-energy range, only a single photon needs to be absorbed to remove the 2s electron, while several photons need to be absorbed to remove the 1s electron. For a meaningful study, we compare emission rates for the 1s and 2s electron after absorption of the same number of photons.

The first study we perform is to investigate how the ratio emission of 1s electron / emission of 2s electron depends on the number of photons required to emit the 1s electron (figure 1). These investigations were carried out at an intensity of 2×10^{13} W/cm^2. Figure 1 shows that when only one or two photons are required to emit the 1s electron, emission of the 1s electron dominates, but when four photons (or more) are required, emission of the 2s electron dominates.

The results in figure 1 are obtained for photon energies at which intermediate resonances have relatively little influence. Their influence only appears at the four-photon level. Hence the straight-line behaviour in figure 1 disappears at N=4. So

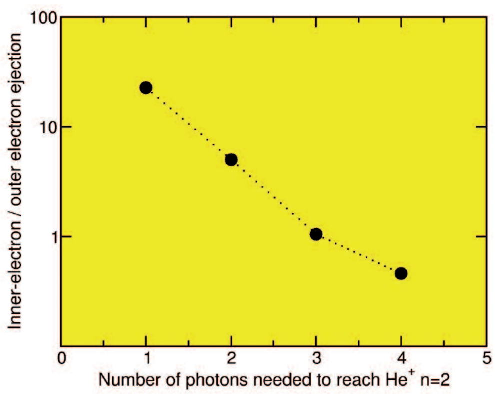

Figure 1. Ratio between emission of the inner electron and emission of the outer electron for the 1s2s ^{1}S state of He as a function of the number of photons needed to emit the inner electron at an intensity of 2×10^{13} W/cm^2.

far, final-state resonances have been neglected, and we will now study whether this neglect is justified.

Figure 2 shows the influence of final-state resonances on the emission rates of the 1s and the 2s electron after absorption of two photons. The results presented in figure 2 are obtained at an intensity of 10^{13} W/cm^2. Figure 2 shows that the final-state resonances influence the ionization rates to a relatively minor extent: the ionization rates typically do not increase by more than one order of magnitude. Extending the study to higher photon energies shows that a substantial increase in the ionization rate is only observed for very narrow resonances. Off resonance, the ionization rates increase by about 10%–25% when the n=3 target states are included. As a consequence, the ratio 1s emission / 2s emission decreases by about 10%. Hence the exclusion of the n=3 target states leads to a reasonable estimate of the ratio between emission of the inner electrion versus emission of the outer electron.

Figure 3 shows the influence of intermediate resonances on the emission rates of the 1s and 2s electrons after absorption of 3 photons. The intermediate resonances are the $2\ell n\ell'$ resonances reached after absorption of 2 photons. In the presented photon energy range, it is also possible to leave He$^+$ in the n=3 states, and these ionization rates are therefore also given in the figure. It can be seen directly that intermediate resonances lead to a much stronger enhancement of the emission rates than the final-state resonances observed in figure 2. Low-lying $2\ell n\ell'$ resonances predominantly leave the residual He$^+$ ion in n=2, while higher resonances leave the residual He$^+$ ion in n=3.

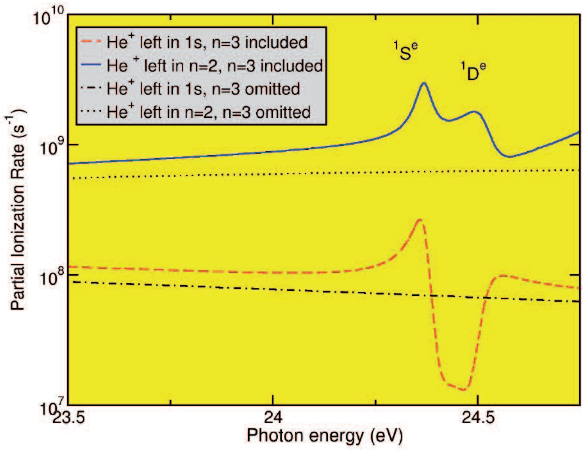

Figure 2. Ionization rates for the 1s2s ^{1}S state of He following the absorption of 2 photons, leaving He$^+$ in either 1s or in the n=2 states. Rates are shown for calculations including the $3\ell n\ell'$ resonances and for calculations omitting these resonances. Calculations are performed at an intensity of 10^{13} W/cm^2.

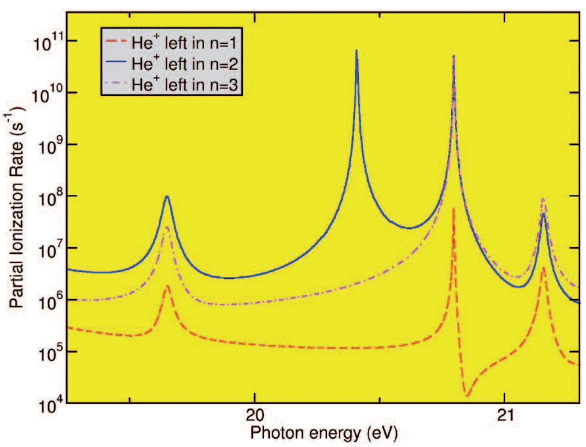

Figure 3. Ionization rates for the 1s2s ^{1}S state of He following the absorption of 3 photons, leaving He$^+$ in 1s, the n=2 states, or the n=3 states. The ionization rates are resonantly enhanced by intermediate $2\ell n\ell'$ states. Calculations are performed at an intensity of 10^{13} W/cm^2.

Two-photon detachment of a 1s electron from Li$^-$

In order to demonstrate that the R-matrix Floquet approach can also be applied to inner-shell emission in more complex systems, we apply the approach to two-photon detachment of the 1s electron from ground-state Li$^-$. Figure 4 shows the rates for two-photon detachment of Li$^-$ leaving the residual Li atom in the autoionizing 1s2s^2 ^{2}S state. The figure further separates the total detachment rate into the two main contributions: emission of an s electron and emission of a d electron. The detachment rates are dominated by emission of s electrons. This can be explained by the angular momentum barrier, which reduces the overlap between the confined 1s orbital in Li$^-$ and continuum d orbitals. Figure 4 shows 2 shape resonances, a ^{1}D resonance at 30.1 eV and a ^{1}S resonance at 30.7 eV. These 1s2s2p^2 resonances coincide with the positions of the 1s2s2p ^{2}P thresholds of Li. The 1s2s2p^2 ^{1}S resonance is important for excitation of the autoionizing 1s2s2p ^{2}P states. Hence the dominating processes for two-photon detachment of a 1s electron from Li$^-$ are (a) direct emission of the 1s electron and (b) excitation of the 1s2s2p^2 ^{1}S resonance. The excitation of this state is aided by 1s^{2}2s^2 – 1s^{2}2p^2 mixing in the Li$^-$ ground state.

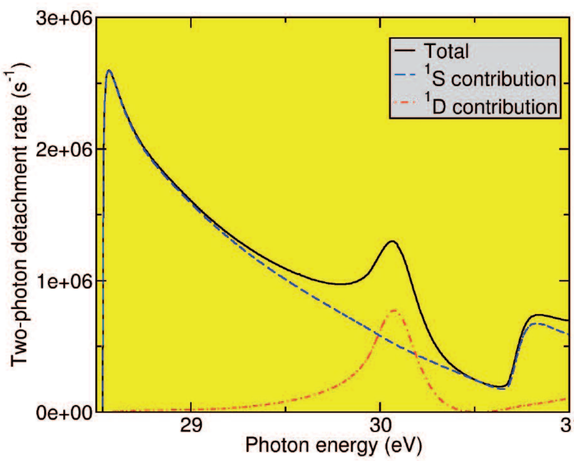

Figure 4. Two-photon detachment rates of the 1s electron from ground-state Li$^-$, leaving Li in the autoionizing 1s2s^2 ^{2}S state at an intensity of 10^{12} W/cm^2. The detachment rates are separated in angular momentum of the outgoing electron.

Conclusions

In the present contribution, we demonstrate that the R-matrix Floquet approach is a valuable technique for the investigation of multiphoton processes involving inner-shell electrons. We have demonstrated for two cases, the 1s2s ^{1}S state in He and the 1s^{2}2s^2 ground states of Li$^-$, that absorption of two photons preferentially leads to the detachment of the inner 1s electron. The study of the He

$1s2s\ ^1S$ state suggests that the ratio 1s emission vs. 2s emission decreases by a factor of approximately 5 for each additional photon required to emit the 1s electron. Intermediate resonances can lead to a significant enhancement of the emission rate for inner-shell electrons, while final-state resonances lead to much smaller enhancements. For the Li^- ground state, we observe that inner-shell emission is dominated by the emission of electrons with very low angular momentum. We characterize the dominant inner-shell emission processes as (a) direct two-photon emission of the 1s electron and (b) excitation of the $1s2s2p^2\ ^1S$ shape resonance.

References

1. K. D. Schultz 2006, private communication
2. H. Wabnitz *et al.,* 2003, *Nature* **420** 482
3. P. G. Burke, P. Francken and C. J. Joachain 1991, *J. Phys. B* **24**, 761
4. M. Madine and H. W. van der Hart 2005, *J. Phys. B* **38**, 3963
5. M. Madine *et al.,* 2006, *J. Phys. B* submitted
6. H. W. van der Hart 2005, *Phys. Rev. Lett.* **95** 153001
7. P. G. Burke and K. A. Berrington 1993, *Atomic and Molecular Processes: an R-matrix approach* (Bristol: Institute of Physics Publishing)
8. N. Berrah *et al.,* 2001, *Phys. Rev. Lett.* **87** 253002