Inner-shell processes in two-photon ionization of Ne^+

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Introduction

Current developments and applications of free-electron laser (FEL) generating laser fields in the VUV-XUV regime allow new physical processes, such as the emission of inner-shell electrons, to be investigated. Several experimental studies on (multiple) ionization of noble gases have been performed [1,2,3]. These studies suggest that new types of ionization processes need to be considered for a full understanding of multiple ionization in intense VUV laser fields.

To understand multiple ionization in detail, we need to take all possible ionization pathways into account. In the VUV regime, this means that we should not only consider emission of outer-shell electrons but also the emission and excitation of inner-shell electrons. The need to account for the response of inner-shell electrons complicates theoretical studies of intense-field ionization. The computational codes need to be able to describe the structure of the atom under investigation accurately. In addition, the number of allowed ionization pathways increases. One code, which allows us to include substantial amounts of atomic structure accurately, is the R-matrix-Floquet code [4,5,6], which has proven to be capable of describing the intense-field response of complex atoms accurately [7,8,9].

In order to investigate the importance of inner-shell excitation and emission processes, we have chosen to study the $1s^22s^22p^5$ ground state of Ne^+. A recent experiment has obtained a generalized cross section for two-photon ionization at a photon energy of 38.4 eV for this ion [2]. An initial theoretical study for this process, including only excitation and emission of the outer 2p electron, obtained a cross section which was a factor of 50 smaller [10]. A possible reason for this huge discrepancy may be the influence of excitation and emission processes involving an inner 2s electron.

Theory

The R-matrix-Floquet approach is a combination of the time-independent R-matrix approach [11] and the Floquet Ansatz. In the Floquet Ansatz, the time-dependent Schrödinger equation is transformed into a set of coupled time-independent equations. The new wave function is described in terms of Floquet blocks, with each Floquet block corresponding with the wave function after absorption or emission of a certain net number of photons. The R-matrix-Floquet approach has already been used to describe a wide variety of processes in the VUV regime, including the two-photon emission of an inner 1s electron from $1s^22s^2$ Li^{-7}), two-photon double ionization of He [8], and multiphoton ionization of Ne and Ar [9].

In the present report we study two-photon ionization of Ne^+. Final states of Ne^{2+} are described using an orbital set containing the 1s, 2s and 2p Hartree-Fock orbitals for the Ne^{2+} ground state [12] and 3s, 3p 3d and 4s orbitals as well as 4p and 4d pseudo-orbitals obtained for electron-impact excitation studies for Ne^{2+} [13]. The Ne^{2+} states are obtained from a configuration-interaction calculation including all possible single excitations of 2s and 2p from $1s^22s^22p^4$, $1s^22s^2p^5$ and $1s^22p^6$. The present calculations include five Ne^{2+} states: $1s^22s^22p^4$ ^3P, ^1D, ^1S, $1s^22s^2p^5$ $^3P^o$ and $^1P^o$.

The basis states for Ne^+ are obtained by combining all Ne^{2+} states with an R-matrix continuum-electron basis set. The basis set also includes all possible double excitations of 2s and 2p from $1s^22s^22p^5$ and $1s^22s^2p^6$ within the set of orbitals, 1s, 2s, 2p, 3s, 3p, 3d, 4s, 4p and 4d, used to determine the Ne^{2+} states. To improve the agreement with experiment, we shift the energy of the ground state and the first excited state of Ne^+ to their experimental values of 41.00 eV and 14.12 eV, respectively.

Within the R-matrix-Floquet calculations, we include all total angular momenta of Ne^+ up to L=5. The inner-region radius is 15 a_0. The Floquet R-matrix is propagated to a distance of at least 55 a_0 before it is matched to the asymptotic solutions. Close to Ne^{2+} thresholds, this propagation distance needs to be increased, and it is extended up to 175 a_0. The Floquet expansion contains 4 absorption blocks and 1 emission block.

Results and discussion

We study two-photon ionization of Ne^+ for photon energies between 25 and 40 eV. At the lower limit, absorption of two–photons starts to approach the $1s^22s^22p^4$ $^1S^e$ threshold. At the upper limit, absorption of a single photon starts to approach the $1s^22s^22p^4$ $^3P^e$ threshold.

Figure 1 shows the two-photon ionization rates for Ne^+ irradiated by laser light with a photon energy between 25 and 40 eV and an intensity of 10^{14} W/cm^2. Several features can be seen in the figure. Due to avoided crossings between intermediate- and final-state resonances with the ground state, the ionization rates are not continuous with photon energy. A detailed explanation of this effect has been given previously [10]. Near a photon energy of 33.2 eV, figure 1 shows a gap in the spectrum. At this photon energy, two-

photon absorption becomes resonant with the $1s^2 2s^2 2p^5\ ^3P^o$ state of Ne^{2+}, and close to the threshold we need to propagate the Floquet R-matrix beyond 175 a_0 to obtain reliable ionization rates.

Very narrow pseudo-resonances, due to the use of pseudo-orbitals, appear in the ionization rates. These are reached after absorption of three or four photons, and show that higher-order processes can affect the ionization rates. We can observe the repetitive features in the Rydberg series leading up to the $^3P^o$ and $^1P^o$ target states.

For photon energies between 25 and 36 eV, figure 1 shows five prominent resonance structures, as well as a Rydberg series converging to the Ne^{2+} $1s^2 2s^2 2p^5\ ^3P^o$ state, just below 33 eV. The resonances at 26.2 eV, 30.3 eV and 34.2 eV are predominantly due to intermediate resonances, whereas the resonances at 31.9 eV and 35.8 eV are predominantly due to $1s^2 2s^2 2p^5(^1P^o)3s/3d$ final-state resonances.

For photon energies above 36 eV, the spectrum in figure 1 becomes more complicated with overlapping Rydberg series converging onto the $1s^2 2s^2 2p^5\ ^1P^o$ threshold at 38.4 eV and the $1s^2 2s^2 2p^4\ ^3P^e$ threshold at 41.0 eV. Figure 1 shows that the extensive calculations give a two-photon ionization rate for Ne^+ in the order of 1.2×10^{12} s^{-1} near 38.4 eV, a factor 3.5 below the ionization rate deduced from the experimental generalized two-photon ionization cross section [2].

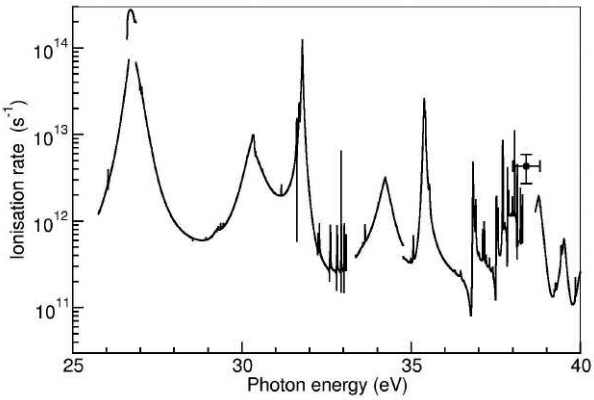

Figure 1. Ionization rates for ground-state Ne^+, irradiated by laser light with a photon energy between 25 and 40 eV and an intensity of 10^{14} W/cm^2. The present ionization rates (solid line) are compared to a rate deduced from the experimental generalised two-photon ionization cross section [2], (solid circle).

To appreciate the importance of this result, we compare in figure 2 the present ionization rates with those obtained from previous work including only emission of the 2p electron [10]. In this previous calculation, only three states of Ne^{2+} were included: $1s^2 2s^2 2p^4\ ^3P$, ^1D and ^1S. Figure 2 shows that the inclusion of Ne^{2+} $1s^2 2s^2 2p^5\ ^3P^o$ and $^1P^o$ final states enhances the ionization rates substantially, with increases of more than one order of magnitude near a photon energy of 38.4 eV. This increase is solely due to additional pathways opening up due to the excitation or emission of a 2s electron. Increases in the ionization rates

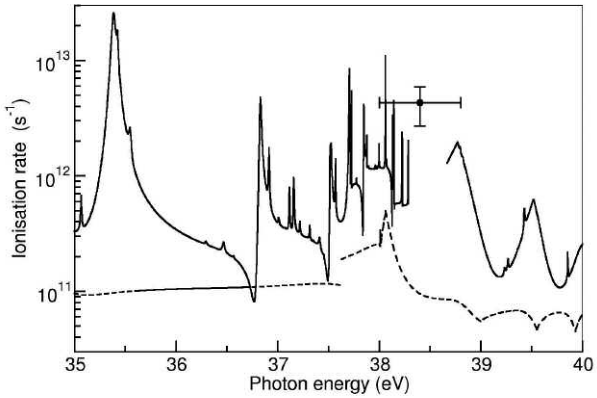

Figure 2. Comparison of ionization rates obtained for ground state Ne^+ irradiated by laser light with photon energy between 35 and 40 eV and intensity I=10^{14} W/cm^2. The five-state calculation, including 2s emission/excitation, (solid line) is compared with a three-state calculation, excluding 2s emission or excitation (dashed line) [7]. Ionization rates are also compared to a rate deduced from the experimental generalised two-photon ionization cross section [2], (solid circle).

are especially noticeable for the Rydberg series leading up to the $1s^2 2s^2 2p^4\ ^3P^e$ threshold at 41.0 eV.

At a photon energy of 38.4 eV, we can compare our ionization rates with experiment. For the three-state calculation, we find a factor of 50 difference between the experimental ionization rate and the theoretical one. For the five-state calculation, a definite ionization rate is difficult to establish as the photon energy coincides closely with the $1s^2 2s^2 2p^5\ ^1P^o$ threshold, and no accurate results can be obtained at the exact experimental photon energy. Within the experimental photon-energy uncertainty, however, we obtain a factor of 3 to 6 difference.

Definite agreement between theory and experiment is difficult to establish. The experimental photon energy has an uncertainty of 0.4 eV and figure 1 shows that the ionization rate is strongly enhanced by resonances. The contribution of resonances depends on intensity I. Ionization rates for the ground state scale with I^2, whereas ionization rates for exited state scale with I. The intensity is constant in the R-matrix Floquet calculations but free-electron laser pulses have intensity fluctuations. The calculations are performed for an intensity of 10^{14} W/cm^2, which is higher than the highest intensity in experiment [2], 3.7×10^{13} W/cm^2. Furthermore, the shape in time and space of the free-electron laser pulse is unknown, and this is needed to transform an ionization yield into a reliable generalized two-photon ionization cross section.

Conclusion

The R-matrix-Floquet approach is a suitable approach to obtain a detailed description of the influence of inner-shell processes on two-photon ionization of Ne^{2+}. These inner-shell processes are found to enhance the ionization rates substantially, in particular by enhancing the ionization rates of intermediate resonances. Hence, an accurate

description of atomic multiphoton ionization processes in the VUV regime requires a detailed description of the atomic structure. The agreement with experiment has improved significantly and is quite satisfactory. However, to further improve the agreement with experiment, we must have a better knowledge of the laser field parameters: the intensity and the shape of the laser field in both space and time are needed.

References

1. H. Wabnitz, A. R. B. de Castro, P. Gürtler, T. Laarmann, W. Laasch, J. Schulz and T. Möller, *Phys. Rev. Lett.* **94**, 023001 (2005).
2. A. A. Sorokin, M. Wellhöfer, V. Bobashev, K. Tiedtke and M. Richter, *Phys. Rev A* **75**, 051402(R) (2007).
3. R. Moshammer *et al.*, *Phys. Rev. Lett*. **98** 203001 (2007).
4. P. G. Burke, P. Francken and C. J. Joachain, *J. Phys. B: At. Mol. Opt. Phys.* **24**, 761 (1991).
5. M. Dörr, M. Terao-Dunseath, J. Purvis, C. J. Noble, P. G. Burke and C. J. Joachain, *J. Phys. B: At. Mol. Opt. Phys.* **25**, 2809 (1992).
6. K. M. Dunseath and M. Terao-Dunseath, *J. Phys. B: At. Mol. Opt. Phys.* **35**, 125-140 (2002).
7. H. W. van der Hart, *Phys. Rev. Lett.* **95**, 153001 (2005).
8. L. Feng and H. W. van der Hart, *J. Phys. B: At. Mol. Opt. Phys*. **36**, L1-L7 (2003).
9. C. McKenna and H. W. van der Hart, *J. Phys. B: At. Mol. Opt. Phys.* **37**, 457 (2004).
10. P. G. Burke and K. A. Berrington, *Atomic and Molecular Processes: An R-matrix Approach* (Bristol: Institute of Physics Publishing, 1993).
11. E. Clementi and C. Roetti, *At. Data. Nucl. Data Tables* **14**, 177-478 (1974).
12. L. Hamonou, H. W. van der Hart, K. M. Dunseath and M. Terao-Dunseath, *J. Phys. B: At. Mol. Opt. Phys.* **41**, 015603 (2007).
13. B. M. McLaughlin and K. L. Bell, *J. Phys. B: At. Mol. Opt. Phys.* **33**, 597 (2000).