Double Photoionization of Strontium

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Abstract. Resonant triple-differential cross-section (TDCS) measurements have been used to study the double photoionization process in strontium. Two sets of measurements were made at the photon energy of the $4p \rightarrow 4d$ resonance. The coplanar geometry was used and the fixed analyser, positioned at $-90^\circ$ to the main axis of polarization of the photons, detected electrons with $\sim 65\%$ of the available excess energy. The mutual angle between the two electrons had a range just short of $90^\circ \rightarrow 270^\circ$. The TDCS exhibit unexpected lobes at a mutual angle of $180^\circ$. Comparison with other measurements made with the same geometry but with different sharings of the available energy indicate that these TDCS all show the unexpected lobe. Some possible explanations for the lobe are considered.

1. Introduction
Double photoionization (DPI), though forbidden by the one electron model of ionization, provides insight into electron correlation inside the atom. Helium, the DPI of which results in a bare nucleus and two electrons in the continuum, has been the focus of many investigations [1]. Alkaline-earth metals are also of interest since their electron configurations are such that there are two $ns$ electrons in the outermost valence shell. These are the two electrons that are removed in the lowest energy DPI process and the remaining electrons may be considered as spectators, playing no direct role in the photoionization process. The focus of the present study is strontium although selected work on other alkaline-earths will be briefly reviewed.

One experimental method employed to study the DPI process is to measure the double to single photoionization ratio and use the single photoionization cross-section to determine the DPI cross-section. This method has been used to study beryllium [2,3] and more recently magnesium [4]. It has been argued that the photon energy dependence of the measured double to single photoionization ratios could be modeled by a suitably scaled ratio for helium [4]. This assumes that the dominant DPI mechanism is a two step one, the second step being that of internal electron impact ionization of the singly charged ion (created in the first step) by a single photoelectron, the impact happening as the initial electron is liberated. However, the scaling model, which worked for beryllium and lithium, did not work for magnesium [4]. The measured double to single ionization ratios for both magnesium and beryllium are in reasonably agreement with the convergent close coupling (CCC) calculations of Khéifets and Bray [5]. However it is important to note that the agreement is significantly poorer close to threshold for magnesium where the calculated ratio is larger than the experimental double to single ionization ratio, the calculated ratio for beryllium also exceeds the experimental one. It is possible that these discrepancies, at excess energies below 10 eV, are associated with the breakdown of the frozen core model adopted for the double photoionization calculations [5].
The DPI process can also be investigated more differentially by measuring triple differential cross-sections (TDCS), which determine the DPI cross-section with respect to the energy of one electron along with the angles of ejection of the two electrons for a particular excess energy. The only TDCS measurements for direct photoionization in alkaline-earth metals have been carried out on calcium [6]. The data differed markedly from similar data for helium, as do the corresponding CCC predictions [5]. However, again it is worth noting that there are discrepancies between the experimental data and predictions which have been ascribed to the fact that the measurements were made at a photon energy that is relatively close to the $3p \rightarrow 3d$ giant resonance.

Due to the low density of target atoms that can be achieved with a metal vapour source and the extremely low cross-sections for the direct DPI process, further alkaline-earth TDCS measurements have been confined to resonant measurements in calcium and strontium [7-10]. Of particular relevance to the present article are the experimental TDCS recorded by West et al on the $4p \rightarrow 4d$ resonance in strontium [9]. These measurements were taken in the coplanar geometry, the two electrons and the plane of polarization vector being in a plane perpendicular to the propagation vector of the incident photon beam. The fixed analyser was positioned at -90° to the major polarization axis of the linearly polarized light. As for the direct calcium TDCS, the strontium TDCS were significantly different from comparable helium measurements. It could be argued that this difference might be due to the resonant nature of the Sr TDCS. Although, Maulbetsch et al have shown that the concepts developed for direct PDI can be successfully used for resonant PDI, specifically TDCS data recorded for Ca in the region of the $3p \rightarrow 3d$ resonance [11]. The Sr TDCS are especially noteworthy due to the presence of a lobe, non-zero, at a mutual emission angle for the two electrons of 180°. This feature violates the selection rules for a transition from a $1S$ state, as is the case for DPI from the Sr $5s^2$ ground state [12]. In the present article further Sr TDCS measurements are considered. These TDCS data were taken at the photon energy of the $4p \rightarrow 4d$ resonance, again with the fixed analyser at -90° to the polarization vector, with the aim of exploring the cause of the unexpected lobe.

2. Experimental details

Two experimental runs were carried out with the same spectrometer [10] using the coplanar geometry, the first at the McPherson beamline 3.2 at the Daresbury SRS and the second at the UE112-PGM1 beamline at BESSY II. The bandpass of the photons at the energy used to acquire the TDCS was 16 meV at BESSY and 25 meV at Daresbury. At BESSY the light was completely linearly polarized, whilst at Daresbury $S_1$ was measured to be 0.7 ± 0.02. $S_1$ describes the degree of linear polarization, parallel over perpendicular with respect to the major polarization direction [9]. The atomic strontium was produced in a cylindrical oven heated to 570° C and directed to the interaction region by 6 converging atomic beams. One of the two 150° spherical sector analysers, the fixed, was positioned at $\theta_1 = -90°$ to the polarization vector of the incident light. The second, rotatable analyser was able to move from approximately $\theta_2 = 0$ to 180°, so that the mutual angle, $\theta_{12}$, had a range just short of 90° to 270°. The analyser bandpass utilized at BESSY was 280 meV compared with 700 meV at Daresbury.

To acquire the TDCS presented here, the analysers were set to accept electrons that shared the available excess energy of 8.52 eV [10], such that the energy of the electron detected by the fixed analyser was 5.51 eV and the energy of the electron detected by the rotatable analyser was 3.01 eV. The rotatable analyser was positioned at 10° intervals across the available angular range and the number of coincidences was monitored by analysing the time difference between coincidence signals from the two analysers using a multi-channel analyser. The true coincidence rate was determined from the coincidence peak, after accounting for random coincidences, and these true coincidences were normalised to the counts in the fixed analyser to account for variations in the incident light flux or the metal vapour density.

3. Results and discussion

Figure 1 shows equivalent TDCS recorded at a) Daresbury and b) BESSY II. The photon energy in both cases was 25.26 eV, the energy of the $4p \rightarrow 4d$ resonance [13]. In both cases the data have been
averaged each side of $\theta_{12} = 180^\circ$ for presentation purposes. The two TDCS are similar in that there are the usual lobes either side of the mutual angle $\theta_{12} = 180^\circ$ and an additional lobe centred at $180^\circ$, which is forbidden by the selection rules [12]. Taking the quantization axis to be along the polarization vector of the photons and considering photo-excitation from an even parity S state, such as the ground state of Sr, with completely linearly polarized light, only odd parity states with $M = 0$ can result. The selection rules forbid electron emission perpendicular to the quantization axis for any $M = 0$ component of an odd parity state, so in the present geometry a node is anticipated in the TDCS at a mutual emission angle of $180^\circ$, for any sharing of the available excess energy. The TDCS shown in figure 1 are consistent with similar earlier data, recorded by West et al at the Daresbury SRS [9], also with the fixed analyser positioned at $-90^\circ$ to the polarization axis of the light. All of the available TDCS, recorded in this geometry, suggest that there is a lobe centred at $180^\circ$, for both equal and unequal sharing of the available excess energy by the two emitted electrons.

**Figure 1.** Sr TDCS recorded at a) the Daresbury SRS and b) BESSY II, at the energy of the $4p \rightarrow 4d$ resonance. The fixed analyser detected 5.51 eV electrons emitted at an angle of $-90^\circ$ (with respect to the polarization direction of the light), whilst the rotatable analyser detected 3.01 eV electrons. $\theta_{12}$ indicates the mutual emission angle of the two electrons. The solid lines shown in panel a) are described in the text.
When comparing the TDCS it is important to consider the effect of the component of the incident photon beam that is polarized at 90° with respect to the main axis of polarization for all the data recorded at the Daresbury SRS, where the parameter \(S_1 < 1\). Light polarized perpendicular to the main polarization axis will generally give rise to peaks in the TDCS at \(\theta_{12} = 180°\). This point is illustrated by the lines included in figure 1a. The solid line is a fit, using the parameterization adopted previously [10], to the TDCS points, that is comprised of a contribution, labeled \(\sigma_o\), corresponding to light polarized along the major axis and a smaller contribution, \(\sigma_y\), resulting from light polarized at 90° to the main polarization axis. Although these two contributions could account for the unexpected lobe for unequal energy sharings, for equal energy sharing back to back emission is forbidden for either polarization direction [12] suggesting that another explanation is required.

Possible explanations for the unexpected lobe have been considered in detail previously [10]. Application of the selection rules [12] indicate that double photoionization from an intermediate state of even parity could give rise to the central lobe at \(\theta_{12} = 180°\) and therefore implies that mechanisms that can access even parity, intermediate states should be considered. This is particularly relevant for the TDCS measured at BESSY II, figure 1b, where \(S_1=1\), precluding any contributions from light polarized at 90° to the major axis of polarization. Perhaps the likeliest mechanism for population of even parity intermediate states that has been considered is the possibility that even parity states can be accessed through mixing of excited states coupled by stray electric fields. It is accepted that the ground state of Sr whilst predominately of the configuration \(4s^24p^65s^2\) contains some components with configurations \(4s^24p^65p^2\) and \(4s^24p^64d^5\) [14]. Excitation from the ground state, including the effects of initial state configuration interaction and allowing for excitation of one or both valence electrons simultaneously with a 4p electron, results in a wealth of structure [14]. In the absence of external fields there is significant mixing between configurations with the same combined occupancy of the 5s, 4p and 4d sub-shells. It is therefore possible that stray fields could mix states with the same combined occupancy but different numbers of 5p electrons and result in intermediate states of even parity. DPI from these even parity states would give rise to a lobe at \(\theta_{12} = 180°\).

However, there is evidence in the data of West et al [9] that stray fields are not the sole reason for the unexpected lobes. Among the TDCS recorded was one in which the excess energy was shared equally by the two electrons and the fixed analyser was position at \(\theta_1 = 0°\) to the polarization axis. As the lobe at \(\theta_{12} = 180°\) is prominent in the equal energy sharing TDCS recorded with the fixed analyser positioned at -90° to the polarization axis, it would be anticipated that there should be some evidence of it, arising from photons polarized perpendicular to the main polarization axis, in the TDCS recorded with the fixed analyser at 0°. There is no evidence of a lobe at \(\theta_{12} = 180°\) in the \(\theta_1 = 0°\) TDCS, suggesting that the unexpected lobe in the \(\theta_1 = -90°\) TDCS is due, at least in part, to something other than mixing due to stray fields. One possibility, that requires further theoretical consideration, is that the resonant nature of the DPI process giving rise to the TDCS in question is significant.

4. Conclusion

Resonant triple-differential cross-section (TDCS) measurements have been performed at the energy of the 4p → 4d resonance in atomic strontium. In particular TDCS in which the electron detected by the rotatable analyser was 3.01 eV have been considered. Measurements recorded at the Daresbury SRS and at BESSY II are similar and exhibit a lobe at a mutual emission angle of \(\theta_{12} = 180°\) that is forbidden by the selection rules for the DPI process. Mechanisms to account for the unexpected feature have been considered. Although double ionization from even parity intermediate states, accessed through mixing caused by stray fields, is perhaps the most likely explanation there is at present no strong theoretical support for this mechanism. It is possible that TDCS measurements made in the presence of a well defined, external electric field could clarify this question. Additionally, mixing due to stray fields will not explain all of the available TDCS in Sr and indicate that theoretical modeling of the DPI would be useful. The present observations together with the discrepancies between calculation and experiment for less differential,
double photoionization cross-section data suggest that there is more to be learned about the DPI process in alkaline-earth atoms.

Acknowledgments

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References

[1] Briggs J S and Schmidt V 2000 J. Phys. B: At. Mol. Opt. Phys. 33 R1-48.
[2] Wehlitz R and Whitfield S B 2001 J. Phys. B: At. Mol. Opt. Phys. 34 L719-725.
[3] Wehlitz R, Lukić D and Bluett J B 2005 Phys. Rev. A. 71 012707.
[4] Wehlitz R, Juranić and Lukić D V 2008 Phys. Rev. A. 78 033428.
[5] Kheifets A S and Bray I 2007 Phys. Rev. A. 75 042703.
[6] Beyer H-J, West J B, Ross K J and De Fanis A 2000 J. Phys. B: At. Mol. Opt. Phys. 33 L767-771.
[7] Ross K J, West J B and Beyer H-J 1997 J. Phys. B: At. Mol. Opt. Phys. 30 L735-740.
[8] Ross K J, West J B, Beyer H-J and De Fanis A 1999 J. Phys. B: At. Mol. Opt. Phys. 32 2927-2934.
[9] West J B, Ross K J, Beyer H-J, De Fanis A and Hamdy H 2001 J. Phys. B: At. Mol. Opt. Phys. 34 4169-4181.
[10] Sheridan P, Grimm M and Sokell E 2008 J. Phys. B: At. Mol. Opt. Phys. 41 165204.
[11] Maulbetsch F, Cooper I L and Dickinson A S 2000 J. Phys. B: At. Mol. Opt. Phys. 33 L119-125.
[12] Maulbetsch F and Briggs J S 1995 J. Phys. B: At. Mol. Opt. Phys. 28 551-564.
[13] Jiménez-Mier J, Caldwell C D, Flemming M G, Whitfield S B and van der Meulen P 1993 Phys. Rev. A. 48 442-451.
[14] Mansfield M W D and Newsom G H 1981 Proc. R. Soc. Lond. A 377 431-448.