

Evidence for the non-statistical population of the $1s2s2p\ ^4P$ metastable state by electron capture in 4 MeV collisions of $B^{3+}(1s2s\ ^3S)$ with H_2 targets



E.P. Benis^{a,*}, S. Doukas^b, T.J.M. Zouros^{c,d}

^a Department of Physics, University of Ioannina, GR 45110 Ioannina, Greece

^b Department of Material Science and Engineering, University of Ioannina, GR 45110 Ioannina, Greece

^c Department of Physics, University of Crete, P.O. Box 2208, GR 71003 Heraklion, Greece

^d Tandem Accelerator Laboratory, INPP, NCSR Demokritos, GR 15310 Ag Paraskevi, Greece

ARTICLE INFO

Article history:

Received 17 June 2015

Received in revised form 30 September 2015

Accepted 2 October 2015

Available online 16 October 2015

Keywords:

Non-statistical population

Electron capture

Cascade feeding

Metastable states

Effective solid angle

ABSTRACT

We have revisited previously published data involving collisions of mixed 4 MeV $B^{3+}(1s^2\ ^1S, 1s2s\ ^3S)$ with H_2 targets (Benis et al., 2002) in search of evidence for the non-statistical production of the $1s2s2p\ ^4P$ long-lived metastable state by single electron capture. Using our recently published method for the accurate determination of the effective solid angle of Auger decaying metastable projectile states in combination with knowledge of the $1s2s\ ^3S$ metastable beam fraction allowed us to determine the ratio $R = ^4P/^2P$ of $1s2s2p$ quartet to doublet production cross sections both formed by electron capture. Our present determination of $R = 2.8 \pm 0.5$, clearly departs from the expected value of $R = 2$ based on statistical spin recoupling arguments and thus provides evidence for the active presence of additional population mechanisms in a new collision system.

© 2015 Elsevier B.V. All rights reserved.

1. Introduction

Zero-degree Auger Projectile Spectroscopy (ZAPS) is a high resolution electron spectroscopy technique that has made considerable progress in obtaining state-selective information about collision dynamics of multiply excited atomic states over the last few decades [1,2]. A recent topic of interest is the determination of the ratio $R = ^4P/^2P$ of $1s2s2p\ ^4P$ to 2P states formed by single electron capture to the $1s2s\ ^3S$ long lived component of ($1s^2\ ^1S, 1s2s\ ^3S$) He-like ion beams in collisions with gas targets. Recent determinations of R for carbon [3,4] and fluorine [5–7], have reported large departures from the expected statistical value of $R = 2$, [8,9] in the process providing interesting explanations as to the possible mechanisms involved such as the dynamic Pauli exchange mechanism [5] and the selective cascade feeding from higher lying states [6,3,7,4].

The reported experimental measurements involve the use of pre-excited $1s2s\ ^3S$ metastable beams, naturally found in the typically mixed state ($1s^2\ ^1S, 1s2s\ ^3S$) He-like beams produced in the accelerator and the production of the metastable $1s2s2p\ ^4P$

whose population must be compared to the population of the prompt $1s2s2p\ ^2P$ states. Consequently, any determination of the ratio R has to pay careful attention both to the correct evaluation of the fraction of He-like ions in the metastable $1s2s\ ^3S$ state as well as to the accurate determination of the effective detection solid angle of the measured Auger yield of the long lived $1s2s2p\ ^4P$ projectile state. Recently, we published [10] a new Monte Carlo type approach (within the SIMION 8.1 [11] charged particle optics simulation environment) to treat the problem of the accurate determination of the effective solid angle of the long-lived 4P state in high resolution Auger electron spectroscopy measurements using a hemispherical deflector analyzer (HDA) with injection lens and position sensitive detector (PSD) [12–15]. Using our newly developed Monte Carlo technique [10] and relying on our older method for accurately determining the metastable ion beam content [16–18], we revisited previously published data involving the collision of 4 MeV $B^{3+}(1s^2\ ^1S, 1s2s\ ^3S)$ with H_2 targets and extracted the ratio of interest R .

2. The experimental setup and measuring method

Details on the experimental setup and the measuring process can be found in [18]. Currently, this particular experimental setup

* Corresponding author.

is located in the Tandem Accelerator Laboratory of NCSR Demokritos in Athens, and is fully operational [19]. In short, in ZAPS, the energetic ion beam passes through a differentially pumped gas cell where collisions take place populating the projectile states. The resulting Auger electrons emitted at zero-degrees with respect to the ion beam are focused/decelerated by the entry lens of the spectrometer and then energy analyzed by the HDA to be recorded on the PSD. For this study the fast B^{3+} He-like beam collides with the H_2 gas targets populating, amongst others, the $1s2s2p$ Li-like ionic states.

The mechanisms involved depend on the initial state of ionic beam. Indeed, the fast B^{3+} beam is delivered by the Tandem accelerator either in: (i) an almost pure ground state $1s^2 1S$ after gas stripping inside the Tandem tank [16] or (ii) a mixed ($1s^2 1S, 1s2s^3S$) state after post-stripping the B^{2+} beam delivered by the Tandem in thin carbon foils. Carrying out the experiments by utilizing both beams is an effective way towards quantitatively separating the processes that correspond to the metastable $1s2s^3S$ part of the beam. Indeed, the measured electron spectra from the ground state beam can in principle be subtracted from the mixed state beam, after appropriately accounting for the fractions of the beam content, resulting in a spectrum corresponding only to the metastable part of the mixed beam. Essential details on this double measurement technique including the necessary cross section calculations of the metastable fraction determination of the beam are given in [18]. We only need to comment here that the ion beam can be delivered also in the $B^{3+}(1s2s^1S)$ state, however, the contribution is small, well within the statistical uncertainties, and thus can be safely neglected [18].

In addition, as mentioned earlier, the spectra of interest include the long-lived metastable $4P$ states which Auger decay along the ion path to and through the spectrometer. Therefore a correction of the solid angle must account for the decay time of the state τ , as well as the variation of the solid angle along the ion path to and through the spectrometer. We recently published a study based on a Monte Carlo type approach utilizing the SIMION 8.1 software for treating the problem of the accurate determination of the resulting *effective* solid angle. The input lens of our HDA does not allow for a straightforward geometrical estimation of the solid angle as previously performed on the two-stage parallel plate spectrometers used in the past [20,21,15]. The detailed mathematical and computational description, which can be found in [10], results in a decay-time dependent correction factor which can be symbolized as $G_\tau = \Delta\Omega_{eff} / \Delta\Omega$.

In ZAPS the double differential cross section derivation from the raw experimental data is obtained by the well known experimental formula given by Eq. 1, applied in most of the detecting systems using analyzers [22]:

$$DDCS_i \equiv \frac{d^2\sigma_i}{d\Omega d\epsilon_i} = \frac{N_{e_i}}{N_i L n \Delta\Omega \Delta\epsilon_i T \eta_i} \quad (1)$$

where N_{e_i} is the number of the recorded electrons in the i -th detection channel on the PSD, N_i the number of ions collected during the measurement, L is the length of the gas cell, n the target gas density (molecules/cm³), $\Delta\Omega$ is the solid angle defined by the mid-distance of the gas cell to the entry aperture of the spectrometer input lens, $\Delta\epsilon_i$ is the energy resolution per channel, T is the transmission of the spectrograph and η_i is the detection efficiency per channel of the PSD. According to the above when this formula involves mixed state ion beams as well as metastable Auger decaying states, as in our case, then it has to be corrected both for the ion beam content (term N_i), as well as for the effective solid angle (term $\Delta\Omega$). Then the correct formula for a metastable Auger state can be expressed as

$$DDCS_{meta} = \left(\frac{1}{G_\tau f}\right) \cdot DDCS \quad (2)$$

while for a prompt state, i.e. a state that decays entirely inside the gas cell,

$$DDCS_{prompt} = \left(\frac{1}{1-f}\right) \cdot DDCS \quad (3)$$

where $f = N_{1s2s^3S} / (N_{1s2s^3S} + N_{1s2s^1S} + N_{1s^2 1S})$ is the metastable ion beam fraction.

3. Results and discussion

The dominant processes involved in collisions of energetic $B^{3+}(1s^2 1S, 1s2s^3S)$ beams with H_2 targets are illustrated in Fig. 1. As shown, all the doublet states can be populated from the ground state via the processes of Resonant Transfer Excitation (RTE) and Non-resonant Transfer Excitation (NTE) [23]. However, these contributions, although vital to the determination of the beam content, can be safely excluded from the final ground state-free spectra. Similarly, the contributions from the $1s2s^1S$ state can be neglected due to the very small fraction of the beam that survives to the target, as explained earlier. Thus, finally only the $1s2s^3S$ contributes to the population of the doubly excited states through the processes of NTE and single electron transfer (T), with the last one also referred to as capture. Moreover, it is documented in the literature that the NTE process is negligible compared to T when using H_2 targets for ion beam energies of 0.5–1 MeV/amu which are of use in our experiments [24]. Thus, the population of the $4P$ and

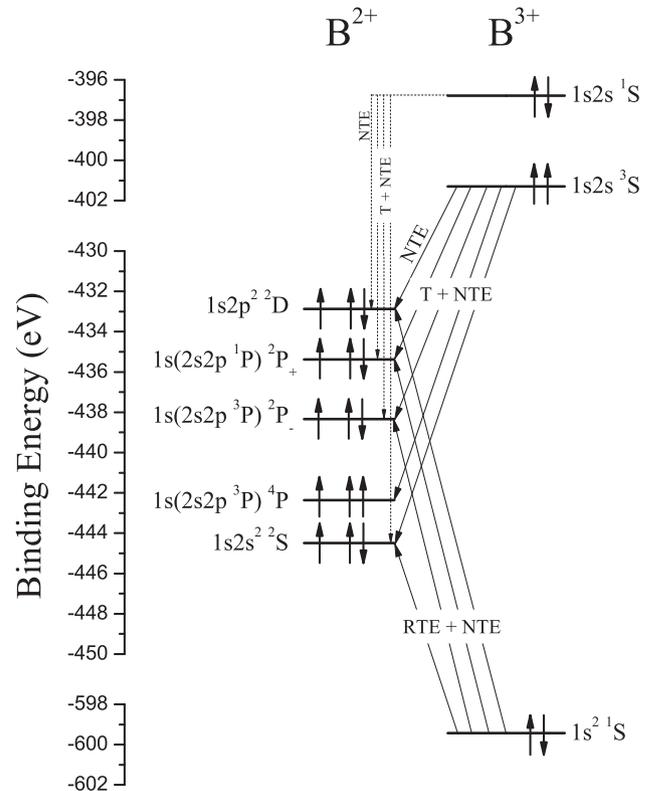


Fig. 1. Energy level diagram showing the dominant mechanisms for the production of the $1s2l2l'(2S+1)L$ doubly excited states formed in collisions of $B^{3+}(1s^2 1S, 1s2s^3S)$ beams with H_2 targets. Also shown are contributions from possible $1s2s^1S$ admixtures that could play a role. For He-like boron beams this component is considered to be negligible.

$2P$ states is achieved primarily via the transfer process to the pure metastable $B^{3+}(1s2s^3S)$ state.

In Fig. 2 we show the electron spectra resulting from collisions of 4 MeV $B^{3+}(1s^2\ ^1S, 1s2s^3S)$ with H_2 targets. As presented in Ref. [18], we were able to obtain pure ground state $B^{3+}(1s^2\ ^1S)$ as well as mixed state $B^{3+}(1s^2\ ^1S, 1s2s^3S)$ beams for the same collision energy. The beam fraction f in the metastable 3S state was determined to have the value of $f = 28 \pm 8\%$.

As can be readily noticed, the two spectra do not have the same energy resolution. This can be understood after considering the processes involved in obtaining the two different content beams. Indeed, in the foil-stripping method (used to produce the mixed-state beam) the uncertainty in the beam energy is higher compared to that of the gas stripping method due to the multiple collisions that the beam suffers in the foil giving rise to energy straggling, as opposed to the primarily single collision conditions established in the gas stripping of the accelerator terminal. Thus, a subtraction of the two spectra after correction for the metastable fraction is not straightforward, but fortunately not absolutely necessary. Since our study involves cross section measurements we can initially obtain the single differential cross sections, $DCS \equiv d\sigma/d\Omega$, after integrating the DDCS over the energy area under the peaks of interest. Then, the resulting DCSs for the $4P$ and $2P$ peaks obtained from the mixed state beams are corrected for the beam metastable fraction. The corresponding DCSs of the ground state beam are subsequently subtracted from the previously corrected mixed-state DCSs, resulting in DCSs ground state-free or equivalently in pure $1s2s^3S$ state DCSs. As a final step the DCS for the $4P$ is corrected for the effective solid angle by dividing it with the correction factor

determined in our case as $G_\tau = 2.33$. The lifetimes we used for these calculations were taken from Refs. [25,26]. Thus, the ratio R for the production cross sections can be written as

$$R = \frac{\frac{1}{\xi_{4P}} \frac{1}{G_\tau} \frac{d\sigma}{d\Omega}(4P)}{\frac{1}{\xi_{2P_+}} \frac{d\sigma}{d\Omega}(2P_+) + \frac{1}{\xi_{2P_-}} \frac{d\sigma}{d\Omega}(2P_-)} \simeq \frac{\frac{1}{G_\tau} \frac{d\sigma}{d\Omega}(4P)}{\frac{d\sigma}{d\Omega}(2P_+) + \frac{d\sigma}{d\Omega}(2P_-)} \quad (4)$$

where a realistic Auger yield value of $\xi \simeq 1$ for all states was finally adopted based on [26]. Moreover, since the angular distributions of the Auger decays from the $4P$ and $2P$ states are similar, R practically corresponds to the ratio of the total production cross sections which based on statistical arguments should result in the value $R = 2$.

According to all of the above, the ratio R was finally determined to have the value $R = 2.8 \pm 0.5$ which clearly departs from the expected statistical value of $R = 2$ [8,9]. Our result thus provides evidence that additional mechanisms (not yet included in the above considerations) are involved. In the literature, the dynamic Pauli exchange mechanism [5] and the selective cascade feeding [6] of the $4P$ state have been proposed as candidates for the enhancement of R . Currently, in the APAPES [19] project underway at the Demokritos Tandem, we are in the process of a systematic experimental and theoretical investigation of the possible mechanisms involved in the enhancement effect. The present experimental results on the value of R for boron, clearly provides further support to this effort and underscores the necessity for further investigation.

As a last step, an additional test providing further credence to the analysis of these results is the determination of the ratio R_{2p} of the doublets $^2P_+/^2P_-$, which under the same spin re-coupling statistical analysis should result in the value of 3. As clearly explained in Ref. [6], these prompt states do not suffer from lifetime solid angle corrections and do not suffer from strong cascade effects or other secondary mechanisms that affect their population. Based on our method and the subsequent corrections for the metastable fraction, we determined this ratio to be $R_{2p} = 2.6 \pm 0.4$ in near agreement with theoretical predictions.

4. Summary and conclusion

We have presented our efforts to experimentally obtain the production cross section ratio $R = ^4P/^2P$ between the quartet and doublet P states formed in collisions of 4 MeV $B^{3+}(1s^2\ ^1S, 1s2s^3S)$ with H_2 targets. This ratio bares the signature of the mechanisms involved in the population of the $1s2s2p\ ^4P$ state, of interest in this study. Using a new approach developed for the calculation of the effective solid angle of long-lived Auger decaying projectile states, we have revisited previously published data from which a value of $R = 2.8 \pm 0.5$ has been determined. In addition, we have also extracted the ratio of the doublets involved, $R_{2p} = ^2P_+/^2P_-$, to have the value of $R_{2p} = 2.6 \pm 0.4$ which is near to the expected result of $R_{2p} = 3$, providing additional support to the correctness of our approach. The observed enhancement in R shows a clear departure from the expected value of $R = 2$, in accordance to earlier evidence, thus adding to the need for further investigations towards a better understanding of the population mechanisms involved.

Acknowledgements

This investigation was partially co-financed by the European Union (European Social Fund-ESF) and Greek national funds through the Operational Program Education and Lifelong Learning of the National Strategic Reference Framework (NSRF)-Research Funding Program: THALES. Investing in knowledge society through the European Social Fund (Grant No. MIS 377289).

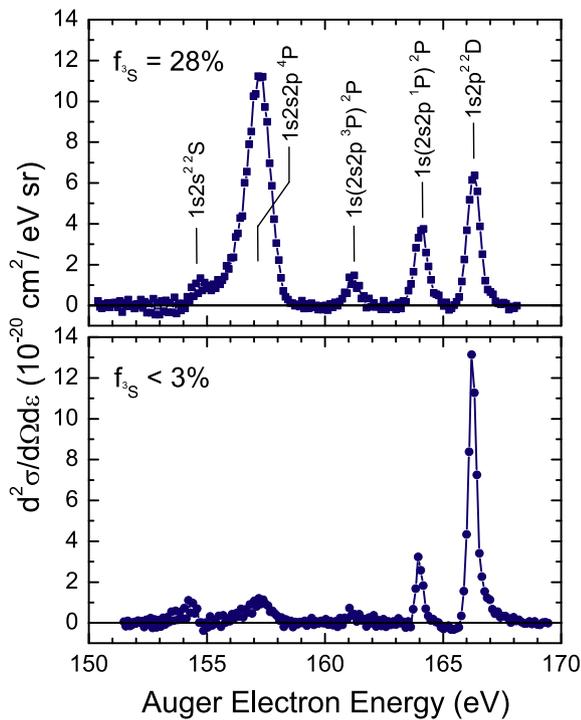


Fig. 2. Experimental electron spectra resulting from collisions of 4 MeV $B^{3+}(1s^2\ ^1S, 1s2s^3S)$ with H_2 targets. [Top] The B^{3+} beam was produced after post-stripping the incident B^{2+} beam in thin carbon foils. The presence of the $4P$ peak, predominantly produced by a $2p$ electron capture to the $1s2s^3S$ ion core, implies a significant metastable component estimated to be $28 \pm 8\%$ [18]. [Bottom] The B^{3+} beam was produced directly from the Tandem accelerator after stripping the initial B^- beam with N_2 gas inside the accelerator terminal. As can be clearly seen, this process leads to a significantly reduced $4P$ peak signifying a practically pure ground-state beam.

References

- [1] T.J.M. Zouros, D.H. Lee, Zero degree Auger electron spectroscopy of projectile ions, in: S.M. Shafroth, J.C. Austin (Eds.), *Accelerator-Based Atomic Physics Techniques and Applications*, American Institute of Physics Conference Series, Woodbury, NY, 1997, pp. 426–479. Ch. 13.
- [2] N. Stolterfoht, R.D. Dubois, R.D. Rivarola, *Electron Emission in Heavy Ion-Atom Collisions*, Springer Series on Atoms and Plasmas, Berlin, 1997.
- [3] D. Strohschein, D. Rohrbein, T. Kirchner, S. Fritzsche, J. Baran, J.A. Tanis, *Phys. Rev. A* 77 (2) (2008) 022706.
- [4] D. Röhrbein, T. Kirchner, S. Fritzsche, *Phys. Rev. A* 81 (4) (2010) 042701.
- [5] J.A. Tanis, A.L. Landers, D.J. Pole, A.S. Alnaser, S. Hossain, T. Kirchner, *Phys. Rev. Lett.* 92 (13) (2004) 133201.
- [6] T.J.M. Zouros, B. Sulik, L. Gulyas, K. Tokesi, *Phys. Rev. A* 77 (5) (2008) 050701.
- [7] T.J.M. Zouros, B. Sulik, L. Gulyas, K. Tokesi, *J. Phys. Conf. Ser.* 163 (1) (2009) 012004.
- [8] E.P. Benis, T.J.M. Zouros, T.W. Gorczyca, A.D. González, P. Richard, *Phys. Rev. A* 69 (2004) 052718.
- [9] E.P. Benis, T.J.M. Zouros, T.W. Gorczyca, A.D. González, P. Richard, *Phys. Rev. A* 73 (2) (2006) 029901.
- [10] S. Doukas, I. Madesis, A. Dimitriou, A. Laoutaris, T.J.M. Zouros, E.P. Benis, *Rev. Sci. Instrum.* 86 (4) (2015) 043111.
- [11] S.I.S. Inc., SIMION 8.1.2.20, Ringoes, NJ, see <http://www.simion.com>, 2005
- [12] E.P. Benis, K. Zaharakis, M.M. Voultzidou, T.J.M. Zouros, M. Stöckli, P. Richard, S. Hagmann, *Nucl. Instrum. Methods Phys. Res. B* 146 (1998) 120–125.
- [13] E.P. Benis, T.J.M. Zouros, P. Richard, *Nucl. Instrum. Methods Phys. Res. B* 154 (1999) 276–280.
- [14] E.P. Benis, T.J.M. Zouros, *Nucl. Instrum. Methods Phys. Res. A* 440 (2000) 462–465.
- [15] E.P. Benis, T.J.M. Zouros, *J. Electron Spectrosc. Relat. Phenom.* 163 (2008) 28–39.
- [16] M. Zamkov, H. Aliabadi, E.P. Benis, P. Richard, H. Tawara, T.J.M. Zouros, *Phys. Rev. A* 64 (2001) 052702.
- [17] M. Zamkov, H. Aliabadi, E.P. Benis, P. Richard, H. Tawara, T.J.M. Zouros, *Phys. Rev. A* 65 (2002) 032705.
- [18] E.P. Benis, M. Zamkov, P. Richard, T.J.M. Zouros, *Phys. Rev. A* 65 (2002) 064701.
- [19] I. Madesis, A. Dimitriou, A. Laoutaris, A. Lagoyannis, M. Axiotis, T. Mertzimekis, M. Andrianis, S. Harissopoulos, E.P. Benis, B. Sulik, I. Valastyn, T.J.M. Zouros, *J. Phys. Conf. Ser.* 583 (2015) 012014.
- [20] D.H. Lee, (Ph.D. dissertation), Kansas State University, (unpublished), 1990.
- [21] D.H. Lee, T.J.M. Zouros, J.M. Sanders, P. Richard, J.M. Anthony, Y.D. Wang, J.H. McGuire, *Phys. Rev. A* 46 (1992) 1374.
- [22] N. Stolterfoht, *Techniques of high resolution Auger electron and x-ray spectroscopy in energetic ion atom collisions*, in: H.O. Lutz, J.S. Briggs, H. Kleinpoppen (Eds.), *Fundamental Processes in Energetic Atomic Collisions*, NATO Advanced Study Institute Series B: Physics, Vol. 103, Plenum Publishing Corporation, New York, 1983, pp. 295–318.
- [23] T.J.M. Zouros, *Resonant transfer and excitation associated with Auger electron emission*, in: W.G. Graham, W. Fritsch, Y. Hahn, J. Tanis (Eds.), *Recombination of Atomic Ions*, NATO Advanced Study Institute Series B: Physics, Vol. 296, Plenum Publishing Corporation, New York, 1992, pp. 271–300.
- [24] D.H. Lee, P. Richard, J.M. Sanders, T.J.M. Zouros, J.L. Shinpaugh, S.L. Varghese, *Phys. Rev. A* 44 (1991) 1636.
- [25] B.F. Davis, K.T. Chung, *Phys. Rev. A* 36 (1987) 1948.
- [26] B.F. Davis, K.T. Chung, *Phys. Rev. A* 39 (1989) 3942–3955.