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Population of the $1s2s(^3S)nl\ ^2L$ states in collisions of mixed-state ($1s^2\ ^1S$, $1s2s\ ^3S$) B^{3+} and C^{4+} ion beams with He and H_2 targets

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Auger KLn lines are observed in high-resolution electron spectra obtained in collisions of mixed-state ($1s^2\ ^1S$, $1s2s\ ^3S$) He-like beams of 4 MeV B^{3+} with H_2 and 6 MeV C^{4+} with He targets. Supporting atomic structure calculations show these lines to correspond to $1s2s(^3S)nl\ ^2L$ doubly excited states, which can be readily populated by electron transfer to the $1s2s\ ^3S$ component of the mixed-state beam. They thus provide indirect evidence for the existence of the corresponding $1s2s(^3S)nl\ ^4L$ KLn quartet states, similarly produced, even though their weak Auger decay does not allow for their direct observation in the electron spectra. These KLn quartet states mostly decay in a cascade chain of strong radiative $E1$ transitions, eventually terminating at the $1s2s2p\ ^4P$ state, which is thus additionally enhanced. An upper limit on the $1s2s2p\ ^4P$ state population due to cascades is obtained by assuming a statistical production of KLn quartet to doublet states followed by a 100% cascade feeding of the $1s2s2p\ ^4P$ state. Our estimated upper limit is supported by our absolute cross section measurements and corresponding three-electron atomic orbital close coupling calculations in progress. Results to date are presented and discussed.

1 | INTRODUCTION

Ion-impact studies continue to play an important role providing insight into fundamental atomic collision processes, such as electron transfer, excitation, and ionization, as well as their combinations, in atomic and molecular few-body systems.[1,2] At the same time, they provide data of relevance for more applied research directions such as the study of controlled thermonuclear fusion, laboratory and astrophysical plasmas, radiation damage of biological tissue, the development of new ion sources, the promotion of new accelerator technology, and the creation of vacuum ultraviolet (vuv) and x-ray lasers.[3–5] In particular, high resolution studies of few-electron ions provide *state-selective* information for theories to be tested to the next order of sophistication and accuracy.[6–11]

Information about these atomic states and their population dynamics in collisions can be obtained from the study of the radiative decay of the excited states (x-ray spectra), as typically done in astrophysics, and of the nonradiative decay (Auger spectra), as typically done in collisions studied in the laboratory. Radiative and Auger decays are complementary relaxation processes providing the necessary information for a thorough study of state population dynamics in collisions. However, for few electron ionic systems of low atomic number ($Z_p < 10$), Auger spectroscopy is preferable due to the much stronger Auger decay rates, A_α, compared with the radiative decay rates, A_x. As a result, the Auger yields $\xi = A_\alpha/(A_\alpha + A_x)$ for such ionic states are typically close to unity, thus favouring the use of Auger electron spectroscopy. Moreover, radiative transitions are primarily allowed only between states obeying dipole selection rules, thus reducing the number of relaxation paths. Then, Auger spectroscopy is the only access to these excited states that are dipole forbidden to radiative decay. A typical example is provided by the $1s2s2p\ ^4P$ state that is spin forbidden to further radiative decay. However, it can Auger decay to the $1s^2$ ground state.

Here, we present measured Auger electron spectra for collisions of mixed-state ($1s^2\ ^1S, 1s2s\ ^3S$) He-like beams of 4 MeV B^{3+} with H_2 and 6 MeV C^{4+} with He targets. Our goal is to search for signatures of the radiative cascade feeding of the $1s2s2p\ ^4P$ from higher lying quartet states. All the $1s2s(^3S)nl\ ^4L$ KLn quartet states are primarily populated by nl electron transfer to the $1s2s\ ^3S$ part of the mixed-state beam. The $1s2s2p\ ^4P$ state, however, acts as a kind of ground state for higher lying radiative transitions, and its population should thus be enhanced by cascade feeding from the $1s2s(^3S)nl\ ^4L$ $(n > 2)$ quartet states. Actually, this cascade feeding mechanism has been proposed as an alternative explanation of the reported enhancement of the $1s2s2p\ ^4P$ population in collisions of few MeV He-like carbon ions with helium targets.[12,13] The results presented here show that such a cascade feeding of the $1s2s2p\ ^4P$ state from higher lying quartet states is viable and consistent with our observed KLn Auger spectra.

2 | EXPERIMENTAL RESULTS AND ANALYSIS

Details of our zero-degree Auger projectile spectroscopy (ZAPS) setup and the measuring process can be found in the literature.[14] Currently, our ZAPS setup is in operation at the Athens 5.5 MV tandem Van de Graaff accelerator laboratory at the National Center for Scientific Research "Demokritos," under the "Atomic Physics with Accelerators: Projectile Electron Spectroscopy" (APAPES) initiative.[15] In short, in ZAPS measurements, the energetic ion beam passes through a differentially pumped gas cell, where collisions take place, populating the projectile states. The resulting projectile Auger electrons, emitted at zero-degrees with respect to the ion beam direction, are focused and preretarded by the spectrometer entry lens, energy analyzed by the hemispherical deflector analyzer and recorded on the two-dimensional position sensitive detector.

In Figure 1 (bottom), we present older double differential cross section (DDCS) data obtained at the tandem accelerator of the J. R. McDonald laboratory at Kansas State University for collisions of mixed-state 4 MeV $B^{3+}(1s^2\ ^1S, 1s2s\ ^3S)$ with H_2 targets. The continuum background has been subtracted and the resulting spectra have been transformed to the projectile rest frame. The B^{3+} ion beams were obtained after stripping the delivered B^{2+} ion beam in thin carbon foils (a technique known as foil post stripping, FPS). In this way, a considerable amount of the long-lived $1s2s\ ^3S$ state is also produced in the beam, as evident by the prominent $1s2s2p\ ^4P$ line in the Auger spectrum, readily produced by direct $2p$ electron transfer to the $1s2s\ ^3S$ ion beam component. An additional small fraction of ions in the $1s2s\ ^1S$ state, produced in a statistical ratio of 1:3 to the $1s2s\ ^3S$ state, is omitted since only a small percentage finally survives to the target area, due to its much shorter lifetime.

The data are accompanied by R-matrix calculations shown in Figure 1 (top).[16] In Figure 1 (bottom), the R-matrix calculations above are compared to the measured

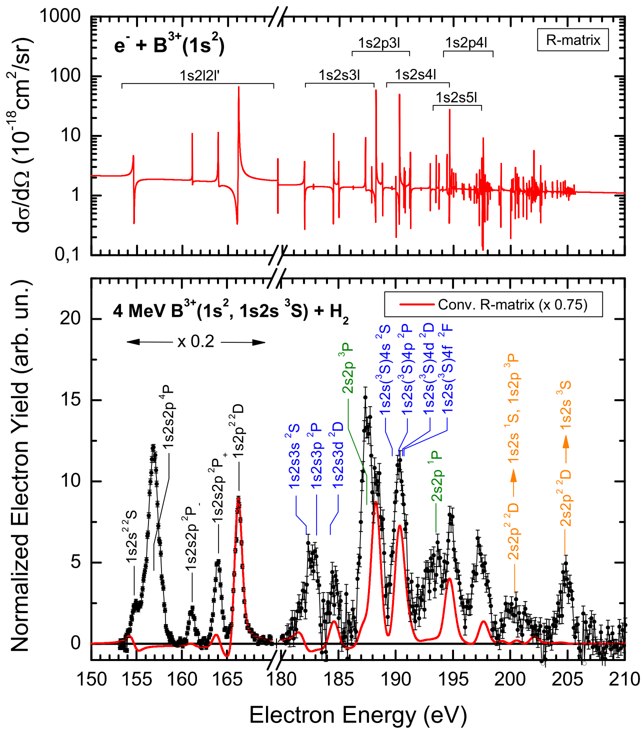

FIGURE 1 (Top) R-matrix calculations of $d\sigma/d\Omega$ for 180° elastic electron scattering from $B^{3+}(1s^2\ ^1S)$ ions plotted as a function of the incident electron energy. (Bottom) Li-like KLn $(n \geq 2)$ Auger spectra obtained in collisions of mixed-state 4 MeV $B^{3+}(1s^2\ ^1S, 1s2s\ ^3S)$ with H_2 targets. Red line: R-matrix calculation (top) after subtraction of the non-resonant continuum, convolution with the target Compton profile and the experimental energy resolution, and then normalized to the $1s2p^2\ ^2D$ state yield. Thus, the rest of the spectrum contributions are from the $1s2s\ ^3S$ beam component. Marked in blue are the states identified by our Cowan code calculations (see text)

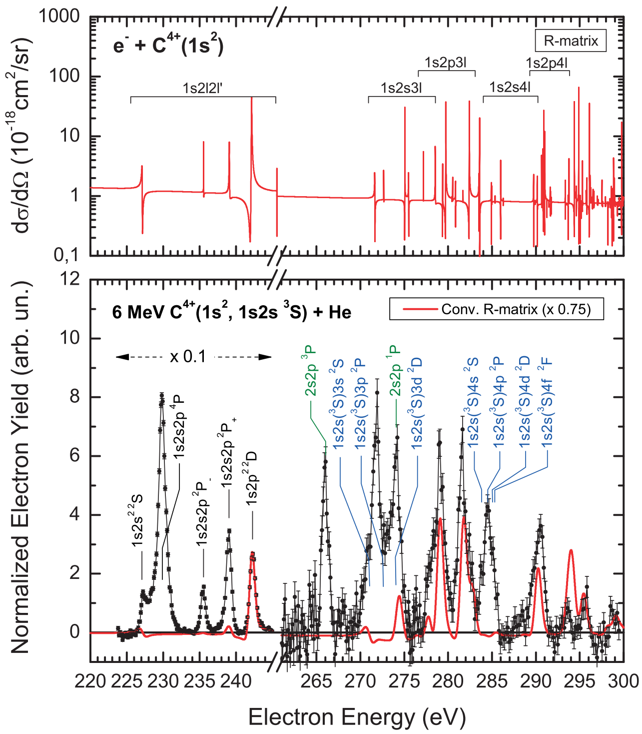

FIGURE 2 Same as in Figure 1, but for collisions of 6 MeV mixed-state $C^{4+}(1s^2\,{}^1S, 1s2s\,{}^3S)$ ion beams with He

normalized yields, after convoluting with the Compton profile of the target, in accordance with the electron scattering model (ESM), resulting in the corresponding DDCS spectra.[16,17] Then, the non-resonant electron scattering continuum was first subtracted from the DDCS R-matrix spectra and subsequently convoluted with the experimental energy resolution of the spectrograph for a direct comparison with the experimental spectra. As a final step, the R-matrix DDCS were further normalized to the experimental $1s2p^2\,{}^2D$ line by multiplying by 0.75,[18] in agreement with fractions reported in the literature for boron.[14,19,20] In this way, contributions from the $1s2s\,{}^3S$ metastable beam component, of interest in this study, are separated from those of the $1s^2\,{}^1S$ ground state. Similar results are presented in Figure 2 for collisions of mixed-state 6 MeV $C^{4+}(1s^2\,{}^1S, 1s2s\,{}^3S)$ beams with He targets. These data were recently obtained with our ZAPS setup at the "Demokritos" tandem accelerator.

3 | DISCUSSION

3.1 | Identification of KLn lines

As can be seen, only the KLL lines in Figures 1 and 2 (top), namely, the $1s2s^2\,{}^2S$, $1s2s2p\,{}^2P_-$ (or $1s(2s2p\,{}^3P)\,{}^2P$), $1s2s2p\,{}^2P_+$ (or $1s(2s2p\,{}^1P)\,{}^2P$) and $1s2p^2\,{}^2D$ states, are clearly identified in the experimental spectra. For the higher energy Auger lines, we performed additional atomic structure calculations based on the Cowan code to help identify the contributions of the various electronic configurations.[21,22] In the case of boron, our Cowan results were compared with the results of the NIST data tables showing an overall agreement better than 0.1%. The contribution of the various $1s2lnl'$ configurations are graphically shown in Figures 1 and 2 (top). These Li-like excited states correspond only to doublet states (^2L), as they are produced from the ground state predominantly through the electron–electron interaction process of resonant transfer and excitation (RTE).[10] They subsequently preferentially decay to the ground state by the much faster Auger transitions rather than the slower $E1$ allowed radiative decays. Thus, any additional radiative feeding between these doublets is minimized and can be safely omitted. It should be mentioned that in ion–atom collisions, the same excited states can also be populated by the process of non-resonant transfer and excitation (NTE), involving nucleus–electron interactions[10] and thus not included in the R-matrix results. Therefore, an additional non-negligible contribution due to NTE is also expected to be present in the observed Auger lines.

By comparing the convoluted R-matrix calculations with the experimental data, the $1s2s\,{}^3S$ contributions to the Li-like KLL doubly excited states can be clearly identified. Indeed, aside from the $1s2s2p\,{}^4P$ state (that can only be produced from the $1s2s\,{}^3S$ component), the contributions to the other KLL states, namely, $1s2s^2\,{}^2S$, $1s2s2p\,{}^2P_-$ and $1s2s2p\,{}^2P_+$, is clearly seen. However, in the KLn ($n > 2$) part of the spectrum, the peaks that are not reproduced by the R-matrix calculations cannot be identified straightforwardly as in the KLL case. After performing atomic structure calculations based on the Cowan code, we were able to identify part of the missing lines as Li-like doubly excited states having configurations of the form $1s2s(^3S)nl\,{}^2L$ and therefore produced from the $1s2s\,{}^3S$ part of the mixed-state He-like beam by direct nl electron transfer. Typical $1s2s(^3S)nl\,{}^2L$ $(n = 3, 4)$ configuration energies are indicated in Figures 1 and 2 (bottom).

3.2 | Identification of hollow states

Aside from the Li-like doubly excited states, the He-like $2s2p\,{}^{1,3}P$ hollow states (not included in the R-matrix calculations) are also identified in the spectra. The $2s2p\,{}^3P$ state is primarily populated by the $1s2s\,{}^3S$ state by direct $1s \to 2p$ electron excitation. The $2s2p\,{}^1P$ state can be populated either from the $1s2s\,{}^1S$ state by direct $1s \to 2p$ excitation or from the $1s^2\,{}^1S$ ground state by double excitation ($1s \to 2s$, $1s \to 2p$; a low probability process). Finally, transfer loss (TL) from either the $1s2s\,{}^1S$ or $1s2s\,{}^3S$ beam components ($2p$ transfer, $1s$ loss) is also possible for both $2s2p\ ^{1,3}P$ states.

In addition, the triply excited $2s2p^2\ ^2D$ hollow state is also clearly evident in the boron spectra in the energy region between 200 and 207 eV. This state is populated predominantly by RTE from the $1s2s\ ^3S$ state and it Auger decays either back to the $1s2s\ ^3S$ state or to the unresolved $1s2s\ ^1S$ and $1s2p\ ^3P$ states. These states are not seen in the carbon spectra (expected at 287 and 282 eV, respectively[23]) possibly due to the use of the He target, having a broader Compton profile compared to hydrogen, that reduces the RTE cross section in this collision energy. The $2s2p^2\ ^2D$ triply excited state, not straightforwardly populated in photoionization experiments, resulted in some of the first tests of R-matrix elastic and inelastic electron scattering off ions with open shells.[23–25]

3.3 | Quartet states and cascade feeding

The only quartet state unambiguously observed in the reported Auger spectra is the $1s2s2p\ ^4P$ state clearly seen in the KLL part. All higher lying $1s2snl\ ^4L$ ($n > 2$) quartet states can be efficiently populated only from the long-lived $1s2s\ ^3S$ component by electron transfer. However, they cannot be easily observed[26] in the Auger spectra due to their very weak Auger decay rates compared with the corresponding much stronger radiative transitions. Therefore, they preferentially decay radiatively to lower lying quartet states via the much stronger $E1$ transitions.[27] In this picture, most of the population of the higher lying quartet states will eventually end up in the $1s2s2p\ ^4P$ state in a cascade feeding chain. The $1s2s2p\ ^4P$ state is forbidden to radiatively decay to the ground state, due to spin selection rules, thus only allowing for its Auger decay to the ground state, albeit with a relatively long lifetime (ns - μs) and thus is termed *metastable*. As a consequence, its detected intensity in the spectrum will depend on the distance between target and spectrometer entry and may appear enhanced or diminished in the Auger spectra.[28] Radiative decay of the $1s2s2p\ ^4P$ state to the ground state $1s^22s\ ^2S$ has been reported in the literature for the higher Z ions of Ar^{16+} but only for the $J = 1/2$ and $J = 3/2$ levels due to the long lifetime of the $J = 5/2$ level.[29]

A possible way to identify the selective enhancement of the $1s2s2p\ ^4P$ state due to cascade feeding is by comparing its yield to that of the corresponding $1s2s2p\ ^2P$ states. As mentioned earlier, the doublet states have much stronger Auger rates, compared with their radiative rates, thus minimizing cascade feeding effects from higher lying doublet states, in contrast to the $1s2snl\ ^4L$ states.[13,30] According to spin statistics and assuming no other effects are involved, $2p$ transfer to the $1s2s\ ^3S$ should result in a ratio of 2:1 for the production of the $1s2s2p\ ^4P$ and $1s2s2p\ ^2P$ states.[16] Therefore, a suggested way to identify cascade feeding is by comparing the ratio of the production cross sections of the ^4P and ^2P states, $R_m = \sigma_{T_{2p}}(^4P)/\sigma_{T_{2p}}(^2P)$, to its spin statistics value of 2. This method had recently been applied, but resulting controversies still await further clarification and additional detailed studying both experimentally and theoretically.[12,13,18,30–32]

Here, we suggest a different way to indirectly identify cascade feeding contributions to the $1s2s2p\ ^4P$ state. Since both $1s2s(^3S)nl$ doublets and quartets can be directly populated by nl electron transfer to the $1s2s(^3S)$ component—there are no selection rules or other symmetries blocking such a process—the observation of the $1s2s(^3S)nl\ ^2L$ doublet states in the Auger spectra should automatically imply the existence of the $1s2s(^3S)nl\ ^4L$ quartet states too. Once the path of the electron transfer to the doublet states is open and unambiguously observed, the path to the quartet states should naturally also be considered. Thus, we could estimate the cascade contributions to the $1s2s2p\ ^4P$ state from the $1s2s(^3S)nl\ ^4L$ quartet states as follows. Indeed, from the comparison between the measured Auger spectra and the R-matrix calculations, we can separate the contributions from the ground state and thus obtain an Auger spectrum corresponding to just the $1s2s\ ^3S$ state. Then, invoking spin statistics, the population of the quartets can be assumed to be double the amount of doublets. However, some care should be exercised in such spin statistic arguments since they are valid only in the single active electron picture. At the next level of sophistication, the transfer of an electron should involve three- or even four-electron wavefunctions corresponding to the number of active electrons of the ion and the target in the collision. In this case, differences in the radial part of the wavefunction for doublet and quartet states and the total spin symmetries should be properly addressed by the theoretical model. In our collaboration, we are currently performing such calculations based on the atomic orbital close coupling (AOCC) approach. Preliminary results indicate that the pure spin statistics ratio of 2 is not reached for the $1s2s2p\ ^{2,4}P$ states or the $1s2snl\ ^{2,4}L$ in general, at least for the collision energies considered here.

Despite whether such spin statistics arguments hold, it is generally accepted that a considerable part of the higher lying quartet states will preferentially radiatively decay, finally ending in the $1s2s2p\ ^4P$ state, thus enhancing its yield. To quantify such contributions, aside from the determination of the cross sections for nl electron transfer to the higher lying quartet states, a detailed cascading model with accurate radiative and Auger decay rates for all the involved states is necessary.[30] However, we may obtain an upper limit estimate to the cascade enhancement, by assuming a 2:1 production of quartets to doublets and a 100% efficient cascade transfer of all higher lying populations to the $1s2s2p\ ^4P$ state. Such an estimate based on the observed production of the $1s2s(^3S)nl\ ^2L$ doubly excited states clearly separated in the spectra of Figures 1 and 2 (bottom), show that the cascade yield enhancement of the $1s2s2p\ ^4P$ state is not larger than a factor of two.

In our collaboration, we are currently investigating, both experimentally and theoretically, the cascade effects in collisions of mixed-state ($1s^2\ ^1S$, $1s2s\ ^3S$) He-like ion beams with gas targets.[13,18,31] Toward this goal, we have developed a two-measurement technique by which we can experimentally separate the contributions of the ground and metastable components,[18] as similarly done here with the use of the R-matrix calculations. In addition, ongoing theoretical developments in the use of three[33,34] and possibly even four active electrons in AOCC calculations to describe all processes in collisions of He-like ions in the $1s2s\ ^3S$ state with He and H targets are presently under way. Preliminary results of this work corroborate the above evidence of non-negligible cascade feeding effects in the production of $1s2s2p\ ^4P$ state and is in qualitative agreement with our experimental and theoretical results to date.

4 | CONCLUSION

In this work, we examined the possibility of indirectly identifying cascade contributions to the $1s2s2p\ ^4P$ state in collisions of mixed-state ($1s^2\ ^1S$, $1s2s\ ^3S$) He-like 4 MeV B^{3+} beams with H_2 and 6 MeV C^{4+} beams with He targets by measuring the emitted full KLn projectile Auger spectra. The identification of $1s2s(^3S)nl\ ^2L$ doublet states populated from the $1s2s\ ^3S$ component of the beam by nl electron transfer in the observed KLn spectra points to the existence of corresponding $1s2s(^3S)nl\ ^4L$ quartet states even though these Auger lines are too weak to be directly observed. Assuming these $1s2s(^3S)nl\ ^4L$ states to be produced according to spin statistics in the ratio of 2:1 with respect to the observed doublets, and a 100% transfer of their population by cascade feeding to the $1s2s2p\ ^4P$ state, an upper limit for the population enhancement of the $1s2s2p\ ^4P$ state was estimated to be not higher than a factor of two. These findings provide additional evidence in support of cascade feeding of the $1s2s2p\ ^4P$ state and are in qualitative agreement with our preliminary experimental and theoretical results.

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