Non-statistical population of $1s^2s^2p^4P_J$ states by charge transfer into multiply charged ions

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Abstract. The experimentally observed enhancement of $(1s^2S_n\ell^2P)$ to $(1s^2S_n\ell^2P)$ populations in collisions of He-like ions with H₂ and He targets is discussed and explained by a state-selective cascade feeding mechanism. Continuum Distorted Wave (CDW) and Classical Trajectory Monte Carlo (CTMC) calculations show that large populations of higher lying $(1s^2S_n\ell^2P)$ states for $\ell = 0, n = 2-8$ can be formed in 0.25-2 MeV/u collisions by direct capture to metastable $F^7^+$ ions. These states subsequently decay through a cascade sequence of radiative or Auger transitions dictated by angular momentum and spin selection rules. All radiative (E1) and Auger transitions for $n \leq 5$ were evaluated using the Cowan package for Hartree-Fock calculations. The $(1s^2S_n\ell^2L)$ quartets were found to Auger decay rapidly to the $1s^2$ level allowing for negligible cascade feeding of lower lying doublets including the $1s^22p^2P_{\pm}$. Alternatively, Auger decay of the $(1s^2S_n\ell^2L)$ quartets is blocked by spin selection rules. Decay can proceed, however, through dipole transitions, but only to other quartets, including the lowest lying $1s^22p^2P_{\pm}$ state, which in this way acts as an “excited ground state” eventually collecting most cascade flux. This selective cascade feeding mechanism thus leads to the non-statistical enhancement of the $1s^22p^2P_{\pm}$ quartet population over those of the $1s^22p^2P_{\pm}$ doublet populations providing an alternative explanation to the recently suggested novel mechanism of dynamical Pauli exchange.

1. Introduction

Cascade feeding of atomic levels has been intensely investigated from the early days of beam-foil spectroscopy [1, 2, 3, 4, 5] to the more recent investigation of wake field effects in fast ions emerging from thin foils [6], the production of hollow atoms by charge transfer in highly charged ion-surface interactions [7], K x-ray emission due to charge exchange between solar wind heavy ions and cometary gases [8], as well as fusion plasma research [9] and references therein. While the process itself is well understood, a detailed cascade feeding investigation is experimentally impractical and theoretically very tedious since a great many transitions are involved. Thus, few complete studies exist and these have been mostly performed at low collision energies. At higher collision energies in the MeV/u range electron capture is known to primarily populate the K- or L-shells and cascade feeding is usually considered to be unimportant.

More relevant to this work dealing with the production of $1s^22p^2P_J$ states in ion-atom collisions, early Auger and x-ray beam-foil investigations of fluorine ions [1, 2] suggested the existence of long-lived Li-like Rydberg ($1sln\ell'^{4}L$) quartet states that should strongly feed lower lying levels. Stolterfoht et al. [4] suggested that the observed strong population enhancement of
the neon 1s2s2p$^4P$ state above statistics, observed in 200 MeV Xe$^{31+}$ collisions with neon, could be due to cascade feeding from such higher-lying quartet states produced in the collision. About a decade later, Bliman et al. [10] found that higher lying (1s2s 3S)nl$^2$P except when L = ℓ) states were produced by direct nl capture in slow 3.8 keV/u collisions of O$^6+$ with He. Anthony et al. [11] and Lee et al. [12, 13] determined the production cross sections $\sigma(4P)$ and $\sigma(2P_\pm)$ for populating the 1s2s2p$^4P$ states in mixed initial state (1s$^2$1s2s$^2$S) He-like C$^4+$ and F$^7+$ collisions with H$_2$ and He using high resolution (zero-degree) Auger electron projectile spectroscopy [14, 15], demonstrating the importance of both electron capture and transfer excitation (TE) mechanisms over the collision energy range of 0.25 – 2 MeV/u. However, all these suggestions of cascade feeding were never backed up by any type of detailed analysis.

More recently, cascade feeding of the 1s2s2p$^4P$ state from higher lying 1s2snl$^2$L states (n > 2) formed by transfer-loss (TL) mechanisms in collisions of ground-state Li-like O$^6+$ (1s2s2s) and F$^8+$ (1s2s2s) ions with He and H$_2$ was shown [16] to largely account for the existence of more than a factor of 10 disagreement between experimentally measured TL cross sections and independent particle model TL calculations with n = 2 [17, 18]. In fact, CDW calculations showed nl transfer into the 1s2s3S levels of O$^6+$ and F$^7+$ for n = 3 – 7 to be as important as capture to just n = 2, even for relatively fast ~1 MeV/u collisions [16]. Thus, it has become clear that cascade feeding can also be important even in faster collisions and needs to be carefully considered where relevant.

In 2004 Tanis et al. [19] reported on the ratio $R=\sigma(4P)/[\sigma(3P_\pm) + \sigma(2P_\pm)]$, where $\sigma(X)$ with X = 1s2s2p$^2$P are the production cross sections of the corresponding F$^8+$ states in collisions of F$^7+$ ions with He and Ne. Auger projectile electron spectroscopy measurements at 1.1 MeV/u found $R=2.9$ for He and $R=2.5$ for Ne. Spin statistics, for purely 2p single electron capture (σ = σ$_{2p}$) to the 1s2s$^2$S long-lived beam component, give R=2 [20, 21]. This seemingly anomalous enhancement of R was interpreted [19] as evidence for a new dynamical 2p capture mechanism named Pauli exchange involving projectile and target electrons with the same spin alignment. In 2008, Tanis et al. [22], using a technique for producing practically pure ground state He-like beams [23, 24, 25], experimentally subtracted the ground state component from the mixed beam spectrum to obtain 1s2l2l$'$ intensities due solely to the C$^4+$ (1s2s$^2$S) component simplifying the theoretical interpretation. However, as in their 2004 article, the suggested Pauli exchange mechanism still remained non-amenable to calculation. Nonetheless, the cascade mechanism was also acknowledged and supporting theoretical calculations suggested that about half of the observed enhancement could indeed be attributed to cascade effects.

In this report, we give a brief account of our cascade feeding analysis resulting in the Auger electron emission from the F$^8+$ (1s2s2p) $^4P$ and $^2P_\pm$ states produced in 0.25 – 2 MeV/u collisions of F$^7+$ ions with He and H$_2$ gas targets as already also presented in Ref. [26] with some additional information. Both our CDW [27] and CTMC [28] results indicate that a substantial number of (1s2s$^2$S)nl$^1$ L Rydberg levels with ℓ = L and n = 2 – 8 are populated by single nl electron transfer to the metastable 1s2s$^2$S ion beam component. An in-depth radiative cascade feeding analysis based on Hartree-Fock calculations of all radiative (E1) and Auger transitions for n ≤ 5 is presented. A cascade mechanism preferentially feeding the 1s2s2p $^4P$ becomes evident. Above the collision energy of 0.7 MeV/u, this satisfactorily accounts for the observed experimental results of Ref. [19] as well as the older measurements of Lee et al. [13, 12] for F$^7+$ collisions on both He and H$_2$. Below 0.7 MeV/u, however, large still unexplained discrepancies persist.

2. Theoretical results
In Fig. 1, CDW total single capture cross sections $\sigma_n$ are shown as a function of the principal quantum number n. The effective ion charge seen by each shell was computed using Slater screening [29] known to improve capture results [27]. Additional CTMC calculations performed at 0.25, 0.5 and 1.1 MeV/u (not shown) were found in good overall agreement with the CDW results for the n-distributions[Fig. 1 (right)] and nl distributions (not shown). Since the CTMC
is a non-perturbative method such agreement provides additional support as to the accuracy of our CDW capture results.

The well-known Cowan Hartree-Fock package [30, 31] was used to calculate all relevant F\(^+\)(1s\(2s\)\(^2\)S) Li-like energy levels, dipole and Auger transition rates for principal quantum number \(n\leq 5\) and \(\ell = 0\), \(n - 1\) with more accurate rates for \(n = 2, 3\) transitions taken from Refs. [32, 33, 34]. In Fig. 2 of Ref. [26], the energy level scheme with transition rates was shown and will not be repeated here. The underlying selective cascade feeding mechanism becomes instantly apparent: the (1s\(2s\)\(^2\)S)\(\ell\)\(^2\)L doublets are found to Auger decay strongly to the K-shell (thick blue transition lines), while the (1s\(2s\)\(^2\)S)\(\ell\)\(^4\)L quartets cannot as they are blocked by spin selection rules. Radiative E1 transitions, however, are readily allowed, but only to lower lying quartets. Thus, radiative branching ratios, BR\(_{rad}\), for transitions between quartets are much larger than for corresponding transitions between doublets, effectively ruling in the strong cascade feeding of the lowest lying 1s\(2s\)2p\(^2\)P states only [26].

Separate quartet and doublet cascade transition matrices were constructed as negligible cross feeding was assumed due to spin conservation. A detailed time-dependent cascade feeding analysis [35] was performed which used the computed capture cross sections \(q(n\ell m_\ell)\) to provide the initial \(t = 0\) (1s\(2s\)\(^2\)S)\(n\ell\)\(^2\)S\(L\)\(^4\)L\(_\jmath\)) population cross sections \(\sigma(nLSJ)\) according to spin and angular momentum coupling statistics requiring \(\sigma(nLSJ) = \frac{(2J+1)}{8\pi(2\ell+1)} \sum_{m_\ell=-\ell}^{\ell} \sigma_{n\ell m_\ell}\). This is seen to give the correct 2:1 ratio of 1s\(2s\)2p\(^4\)P to 1s\(2s\)2p\(^2\)P configuration populations [21], i.e. \(\sum_J \sigma(n = 2, L = 1, S = 3/2, J)/\sum_J \sigma(n = 2, L = 1, S = 1/2, J) = 2\). We note that a mixed (1s\(2s\)\(^2\)S, 1s\(^2\)) component He-like beam also allows for the population of the 1s\(2s\)2p\(^2\)P\(_\pm\) levels by non-negligible Transfer-Excitation (TE) from the F\(^+\)(1s\(^2\)) ground state [13, 12] included here.

In Fig. 2 (left) our results are compared to the 0\(^\circ\) Auger electron emission single differential cross sections (SDCS) measurements of Lee et al. [13, 12] based on the metastable beam fractions determined by Teresawa et al. [36]. Good overall agreement is observed between theory and experiment above about 0.7 MeV/u. The Auger Resonant Transfer Excitation (RTEA) and Non-resonant Transfer Excitation (NTEA) contribution assessment is also included. For the H\(_2\) target, the distinct hump in the doublet SDCS due to RTEA [13, 12] is very nicely reproduced by our calculations [37, 15]. The energy dependence of the data is well reproduced even on an absolute scale when cascade contributions are included, while theory clearly underestimates the
experiment without the cascade contributions. Furthermore, the cascade contributions are seen to be needed only in the case of the $1s2s2p^4P$ results consistent with our model. With decreasing collision energy below 0.7 MeV/u, cascade feeding becomes increasingly more intense and even the $1s2s2p^2P_{\pm}$ levels start to receive some cascade contributions (see also Fig. 3 of Ref. [26]). Our cascade analysis for $n \leq 5$ seriously underestimates both quartet and doublet production at low collision energies for reasons not yet understood. The disagreement is worse for doublert from collisions with $H_2$ targets, seemingly ruling out non-resonant transfer excitation (NTE) as a possible cause, since this is known to be much weaker for $H_2$ than for He [37].

To estimate additional possible contribution from higher lying levels with $n > 5$, an upper limit CDW estimate on the Auger SDCS is also provided in Fig. 2. It ideally assumes all capture to $n = 2 – 8$ levels ends up directly into the $1s2s2p^4P$ or $^2P_{\pm}$ states. In reality, cascade mechanisms will transfer this population to the lower-lying levels with decreasing efficiency as the number of steps needed is increased (cascade order [5]). Below 0.7 MeV/u, the measured SDCS for the $^4P$ are seen to lie above this limit, while for the $^2P_{\pm}$, they lie mostly below. Consequently, the theoretically available direct capture population seems to be clearly insufficient in the case of the $^4P$ levels. In the case of the $^2P_{\pm}$ levels this is not as clear. Either higher capture populations

"Figure 2. (Color on line) Absolute 0° Auger emission SDCS for the $1s2s2p^4P$ and the sum of $1s2s2p^2P_{\pm}$ levels for $H_2$ and He (left) and their ratio $R_e$ (right). Data points from the measurements of Lee et al [13, 12]. Lines and triangles: Theoretical CDW and CTMC results, respectively. Solid red lines and filled triangles include cascade feeding from $(1s2s^3S)n\ell$ with $n = 3 – 5$, while dashed blue lines and open triangles do not ($n = 2$ only). Transfer-Excitation (TE) Auger contributions (both RTEA and NTEA) to the $^2P_{\pm}$ from the $1s^2$ are also shown. TE contributions to the $^4P$ are assumed negligible [12]. An upper limit estimate to capture into the sum of $n = 2 – 8$ levels is also shown. (Right) Ratio $R_e$ of experimental and theoretical Auger SDCS shown on left. Expected theoretical spin statistics ratio assuming isotropy is 1.825 [26]."
are needed under the present inefficient cascade transfer to the $^2P_\pm$, or a much more efficient cascade transfer mechanism with the present calculated capture populations. Clearly, at low energy more $^2P$ intensity is missing than $^4P$ as is evident from the overestimation of their theoretically calculated ratio $R_e$ (right).

To further investigate this puzzling state of affairs we suggest the following possibilities: (a) “cross feeding” between the quartet and doublet populations, considered here to be negligible to first-order, could transfer available capture flux from doublets to quartets this way resolving both puzzles, (b) low energy capture could be more intense than what has been calculated, (c) calculated low energy $\ell$-distributions could be wrong particularly for the highest $\ell$-levels (the Yrast levels) known to contribute most to cascade feeding [5], (d) de-alignment effects at 0° observation [38, 39] need to be carefully considered given possible redistribution of magnetic sub-state populations due to cascade feeding [40, 41]. When included just for 2p capture they were found to affect Auger yields within a factor of 2, particularly for He. Best overall agreement, however, was found assuming isotropy as shown here, (e) A new mechanism further enhancing capture cross sections at the lowest collision energies might be needed. However, such a mechanism would need to enhance both the $^2P$ and $^2P_\pm$ population levels and not just the $^4P$ as proposed by Tanis et al. since both seem to be in need of additional capture flux as already seen in Fig. 2. Furthermore, as shown in the recent $C^{4+}$ results on Pauli exchange [22], the experimental ratio $R$ increases from 6 to 8 as the collision energy increases from 0.5 to 1 MeV/u. This is particularly puzzling given that most exchange interactions usually decrease in strength as the required interaction time is also correspondingly decreased [17], (f) the measured cross sections of Lee et al. might be too large at the lowest collision energies if computed with the wrong metastable fraction since this was not measured under the same conditions, but taken from the older measurements of Teresawa et al. [36].

The ratio of quartet to doublet SDCS is plotted in Fig. 2 (right). $R_e$ with both cascade and TE corrections (red line) is clearly in better agreement with the data than without the cascade corrections (blue line). For H$_2$ agreement is excellent, even reproducing the minimum around 1.1 MeV/u due to RTE, while for He it is also as good. However, below 0.7 MeV/u, the effect of the observed disagreement in the absolute values of the computed SDCS is clearly evident.

3. Summary and Conclusions

In conclusion, we have shown through extensive CDW and supporting CTMC single capture calculations that $s^2sn\ell$ levels of both quartets and doublets can be expected to be strongly populated for $n \lesssim 8$ in collisions $F^{7+}(s^2s^3S)$ ions with H$_2$ and He targets. These states subsequently decay through a cascade sequence of radiative or Auger transitions dictated by angular momentum and spin selection rules. A detailed time-dependent cascade feeding calculation including all radiative (E1) and Auger transitions for $n \leq 5$ shows the $(s^2s^3S)n\ell^2L$ doublets to Auger decay rapidly to the $1s^2$ level allowing for negligible cascade feeding of lower lying doublets including the $1s^2s^2p^2P_\pm$. Alternatively, Auger decay of the $(s^2s^3S)n\ell^4L$ quartets is blocked by spin selection rules, with decay proceeding through intense dipole transitions, but only to other quartets, including the lowest lying $1s^2s^2p^4P$ state. This results in the strong enhancement of the $1s^2s^2p^4P$ level populations relative to the similarly configured $1s^2s^2p^2P_\pm$ doublets. Our calculations are in excellent agreement with experimental results when cascade contributions are included over the energy range of 0.7-2 MeV/u. However, at collision energies below 0.7 MeV/u, where cascade feeding is most intense, strong disagreement still persists. This is particularly puzzling given the good agreement above 0.7 MeV/u. Some suggestions for further investigation are proposed. Clearly more theoretical work is needed, particularly at the lower collision energies, to arrive at a more satisfactory explanation. Additionally, it would also be interesting to experimentally investigate the isoelectronic behavior of $R$.  


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