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Selective enhancement of $1s2s2p$ ^4P_J metastable states populated by cascades in single-electron transfer collisions of $F^{7+}(1s^2/1s2s\ ^3S)$ ions with He and H_2 targets

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A mechanism for the *selective* population of $1s2s2p$ ^4P_J states by electron capture in energetic collisions of $F^{7+}(1s2s\ ^3S)$ ions with H_2 and He is elucidated. Capture calculations indicate $(1s2s\ ^3S)nl\ ^{2,4}L$ doublet and quartet levels to be approximately evenly populated for $n=2-5$. Following capture the doublets Auger decay strongly to the $1s^2$ ground state allowing for negligible feeding of other lower-lying doublets by radiative transitions. The quartets, however, find this decay channel blocked by spin conservation and instead radiatively cascade through lower lying *quartets*, eventually strongly populating the lowest-lying $1s2s2p$ ^4P_J levels in agreement with older experimental results for collision energies above 0.7 MeV/u.

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Over the last decades considerable progress has been made in obtaining information on both the atomic structure and dynamics of multiply excited states using high resolution optical [1] and Auger electron spectroscopy [2,3]. This interest has been generated to a large degree in the fields of plasma physics, thermonuclear fusion research, and astrophysics where the collisional properties of highly stripped ions play an important role.

The determination of highly accurate excitation energies, transition rates, and lifetimes combined with production cross section information obtained from line intensity measurements lead to a better overall understanding of the dominant processes at play. Very often, however, the dominant mechanism is complemented by additional processes usually considered of secondary importance. Here, we call attention to such a secondary mechanism and show how it can lead to the strong *selective* enhancement of a particular multiply excited state thus affecting the overall interpretation of its production. This mechanism can be quite general, minimally requiring the availability of a population of *highly excited* states, some of which need to be *metastable*. A detailed analysis is provided by an example involving the production of highly excited $(1s2s\ ^3S)nl$ doublet ^2L and quartet ^4L states (with $L=l$) in collisions of F^{7+} ions with H_2 and He targets.

The dominant process leading to the production of the $(1s2s\ ^3S)nl\ ^{2,4}L$ states is direct single nl electron capture into the $F^{7+}(1s2s\ ^3S)$ component of the ion beam. In low-Z He-like ion beams such as F^{7+}, it is well known that due to the long lifetimes of the $(1s2s\ ^3S)$ states appreciable admixtures of this state can exist together with the $1s^2$ ground state in the collision. While the doublet ^2L states have mostly femtosecond lifetimes decaying promptly, the ^4L states are 10–1000 times longer lived. This metastability arises largely from spin conservation rules forbidding the conversion of a spin quartet to a doublet by either radiative or Auger deexcitation. This rule eliminates "cross feeding" between the two series, which thus are assumed to evolve independently in time.

The lowest-lying level of the quartet $(1s2s\ ^3S)nl\ ^4L$ series, the $1s2s2p$ ^4P state, is the most metastable with lifetimes in the nanosecond range. In the absence of strong spin-orbit interactions, as in the case of low-Z ions, the $1s2s2p$ ^4P_J states can only decay to the ground state through much weaker spin-spin interactions or higher-multipole radiative transitions [4]. It thus acts as a kind of "excited" ground state collecting the population of all higher-lying quartet states through a chain of radiative transitions mediated by other quartets, eventually leading to its enhanced production. It is important to also realize that the lowest-lying levels of the doublet $(1s2s\ ^3S)nl\ ^2L$ series, the $1s2s2p$ $^2P_\pm$ states, will be relatively much less affected by the cascade-feeding process since most of the available population is quickly siphoned away via the much stronger direct Auger and/or radiative decays to the ground state. Thus, the ^4P states become selectively enhanced relative to the $^2P_\pm$ states as demonstrated in this analysis.

The mechanisms for the production of the $1s2s2p$ ^4P states in low-Z ion-atom collisions have been of continuous interest. However, even though the importance of cascade feeding mechanisms have been mentioned or even discussed to some extent [5–10], their significance has gone largely unnoticed, probably due to the lack of detailed supporting evidence. More recently, Tanis *et al.* [11] invoked a process named the *Pauli exchange interaction* to explain the observed nonstatistical enhancement of the ratio of $1s2s2p$ ^4P to $1s2s2p$ $^2P_\pm$ states in the collision of F^{7+} ions with He targets. Alternatively, Zouros *et al.* [12] recognized the importance of the cascade-feeding mechanism in resolving the long standing discrepancy of more than a factor of 10 between theory and experiment in the production of the $1s2s2p$ ^4P state by transfer loss in collision of Li-like $O^{5+}(1s^22s)$ and $F^{6+}(1s^22s)$ ions with He and H_2. However, they left unanswered the important question as to why the additionally observed $1s2s2p$ $^2P_\pm$ states were not also similarly enhanced. The proposed selective cascade-feeding mechanism provides a clear answer to both cases. Strohschein *et al.* [13] very recently have reported results on the C^{4+}+He system in which the nonstatistical enhancement of the ^4P is attributed equally to Pauli exchange and to feeding by cascades in partial agreement with results reported here.

In this Rapid Communication, we investigate theoretically

Auger electron emission from the $F^{6+}(1s2s2p)\ ^4P$ and $^2P_{\pm}$ states produced in 0.25–2 MeV/u collisions of F^{7+} ions with He and H_2 gas targets. Our results are compared to the extensive zero-degree Auger projectile spectroscopy [3] data of Lee *et al.* [5]. Our detailed continuum distorted wave (CDW) [14] and classical trajectory Monte Carlo (CTMC) [15] results indicate that a substantial number of $1s2snl\ ^{2,4}L$ Rydberg levels with $n=2-7$ are populated by single nl electron transfer to the $1s2s\ ^3S$ ion beam component. An in-depth analysis of the radiative feeding by cascades based on Hartree-Fock calculations of all radiative ($E1$) and Auger transitions for $n\leq 5$ is presented, definitively demonstrating the importance and selectivity of the cascade-feeding mechanism.

In Fig. 1, total capture cross sections σ_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 [16] known to improve capture results [14]. Thus, an ion charge of $q=7.8$ was assumed for nl electron capture into $2s,2p$ orbitals, $q=7.15$ for $3s,3p$, and $q=7$ for $3d$ and higher. σ_n is seen to be important and rather evenly distributed over the entire $n=2-7$ range even at the highest collision energies where *inner* shell capture dominates. CTMC are nonperturbative calculations, typically applicable at lower collision energies [17]. Applied at 0.25, 0.5, and 1.1 MeV/u in good overall agreement with the CDW, they provide additional support as to the accuracy of the capture cross sections.

We use the well-known COWAN Hartree-Fock package [18,19] to calculate all relevant $F^{6+}(1s2snl\ ^{2,4}L_J)$ Li-like energy levels, including dipole and Auger transition rates for principal quantum number $2\leq n\leq 5$ and $l=0,n-1$ with more accurate rates for $n=2,3$ transitions from Refs. [4,20]. In Fig. 2 the energy level scheme with transition rates is shown. The underlying selective cascade-feeding mechanism becomes instantly apparent. The $(1s2s\ ^3S)nl\ ^2L$ doublets are

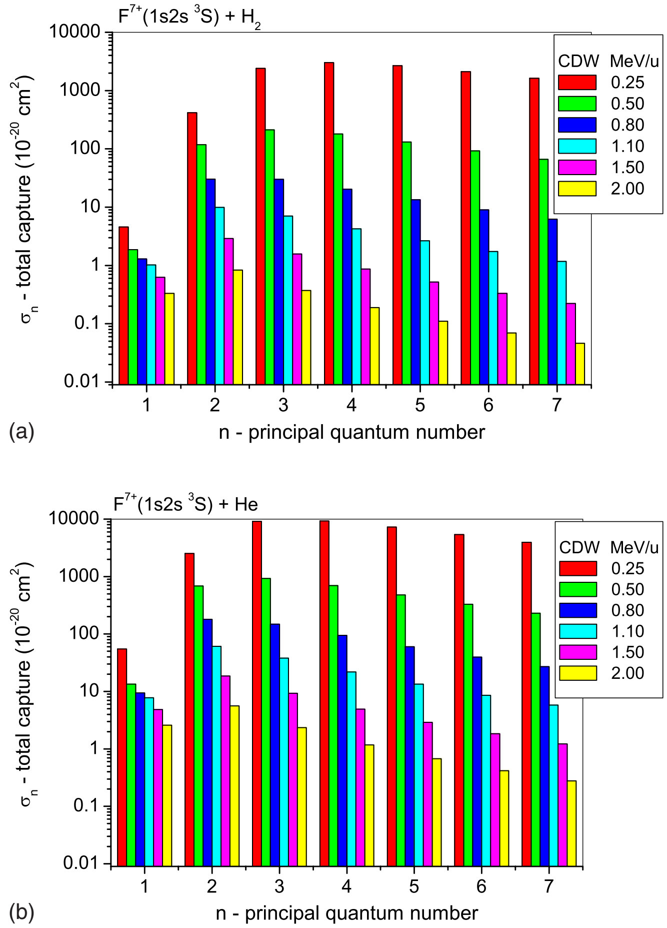

FIG. 1. (Color online) n distribution of CDW cross sections σ_n for single capture into the $1s2s\ ^3S$ state of F^{7+} in collisions with H_2 (top) and He (bottom) for different collision energies.

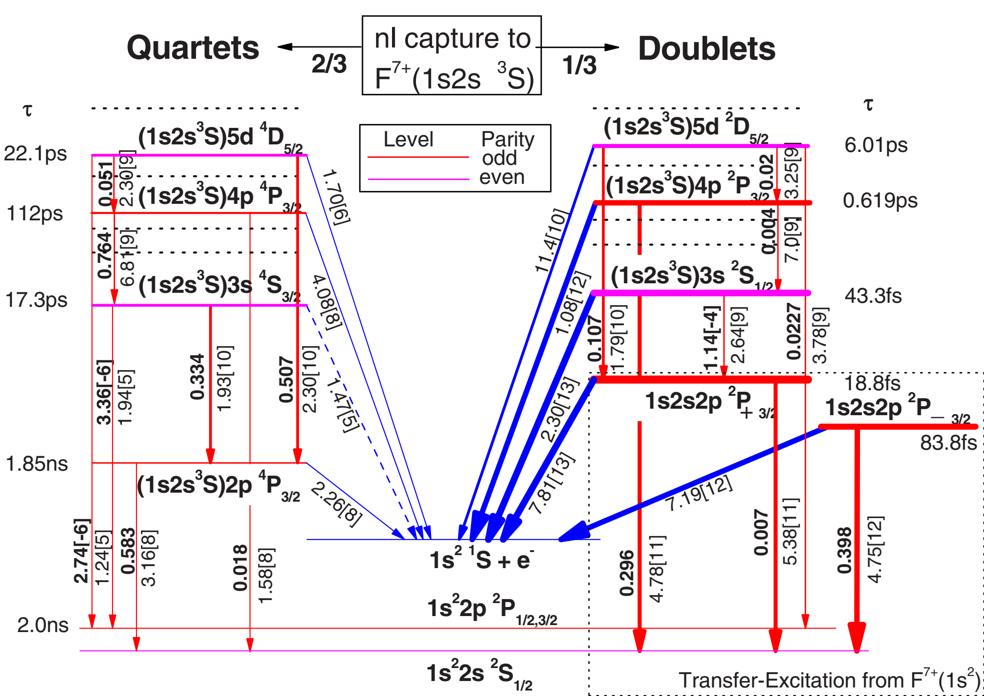

FIG. 2. (Color online) Li-like quartet and doublet $F^{6+}(1s2snl)$ energy level scheme (not to scale) resulting from single electron nl capture to $F^{7+}(1s2s\ ^3S)$. Only a few representative levels are indicated for clarity. Arrows represent transitions with widths roughly proportional to their strength [radiative $E1$ (vertical red lines) and Auger (slanted blue lines)]. Rates (in s^{-1}) are given to the right of the arrows (the quantity in square brackets indicates power of 10), while radiative transition branching ratios, $\mathcal{B}_{rad}$, are given in bold to their left. Also indicated are total lifetimes τ and dashed arrows for Coulomb forbidden transitions.

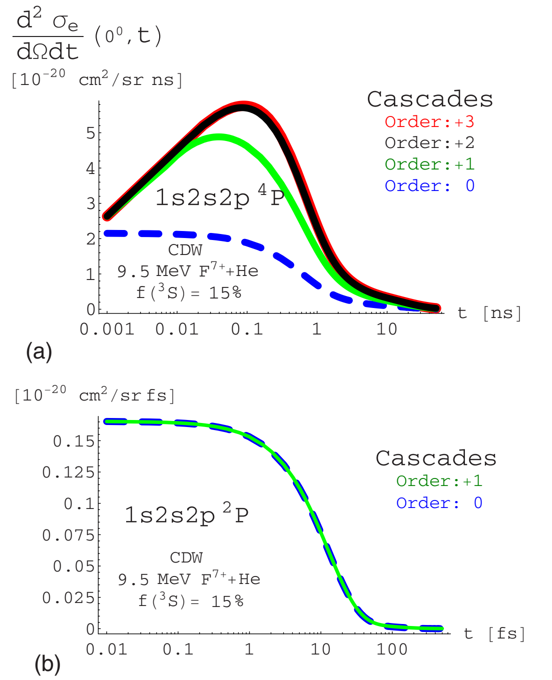

FIG. 3. (Color online) CDW 0° Auger emission SDCS time dependence from the $1s2s2p\ ^4P$ state (top) and $1s2s2p\ ^2P_{\pm}$ states (bottom) in 0.50 MeV/u F^{7+}+He collisions. Lines (in increasing order): Cascades of 0th order (no cascades) (blue dashed), 1st order (green), 2nd order (black), 3rd order (red). Substantial feeding of the $1s2s2p\ ^4P$ and negligible feeding by cascades of the $1s2s2p\ ^2P_{\pm}$ states is observed. Note the difference in time scales, the $1s2s2p\ ^2P_{\pm}$ levels being prompt, while the $1s2s2p\ ^4P$ metastable. Transfer excitation has also been included.

found to Auger decay strongly to the K shell (thick slanted blue transition lines), while the $(1s2s\ ^3S)nl\ ^4L$ 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, $\mathcal{B}_{rad}$, for transitions between quartets are much larger than for corresponding transitions between doublets, effectively resulting in the strong feeding by cascades of the lowest lying $1s2s2p\ ^4P$ states only.

Separate quartet and doublet cascade transition matrices were constructed and a detailed time-dependent analysis [21] of the cascade-feeding process was performed using the computed capture cross sections to provide the initial $t=0$ populations. The individual $(1s2s\ ^3S)nl\ ^{2S+1}L_J$ initial level populations were determined by spin and angular momentum coupling statistics which lead to the well-known 2:1 ratio of ^4L to ^2L configuration populations [22]. An example of the computed time dependence is shown in Fig. 3 providing quantitative support for the proposed selective cascade-feeding mechanism. We note that a mixed $(1s^2/1s2s\ ^3S)$ component He-like beam also allows for the population of the $1s2s2p\ ^2P_{\pm}$ levels by *non-negligible* transfer excitation (TE) from the $F^{7+}(1s^2)$ ground state [5] included here.

In Fig. 4 our results are compared to the 0° Auger electron emission single differential cross sections (SDCS) measurements of Lee *et al.* [5] based on the metastable beam fractions determined by Teresawa *et al.* [23]. It is not clear to

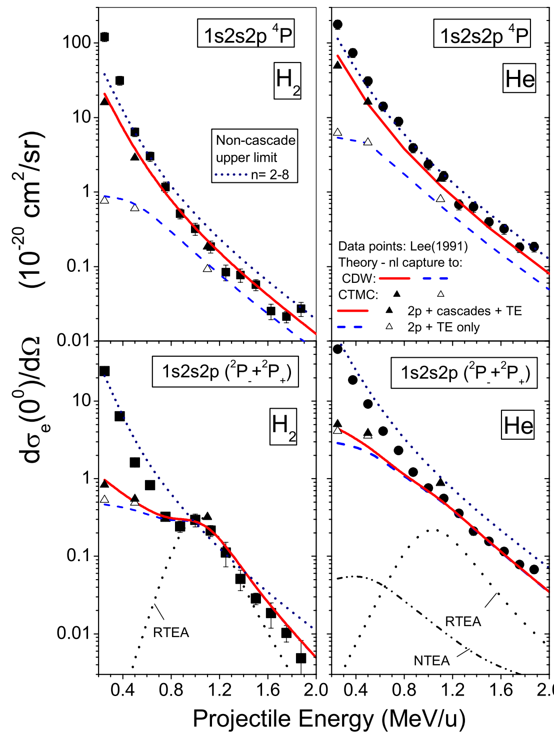

FIG. 4. (Color online) Data points: Absolute 0° Auger emission SDCS for the $1s2s2p\ ^4P$ (top) and the sum of $1s2s2p\ ^2P_{\pm}$ (bottom) levels for H_2 (left) and He (right) (from [5]). Theory: Lines (CDW) and triangles (CTMC). Red continuous lines and filled triangles include feeding by cascades from $(1s2s\ ^3S)nl$ with $n=3-5$, while dashed blue lines and open triangles do not ($n=2$ only). TE (RTE and NTE) Auger contributions to the $^2P_{\pm}$ from the $1s^2$ are also shown. There is no TE contribution to the ^4P. An upper limit estimate to capture into the sum of $n=2-8$ levels is also given (densely dotted line; see text).

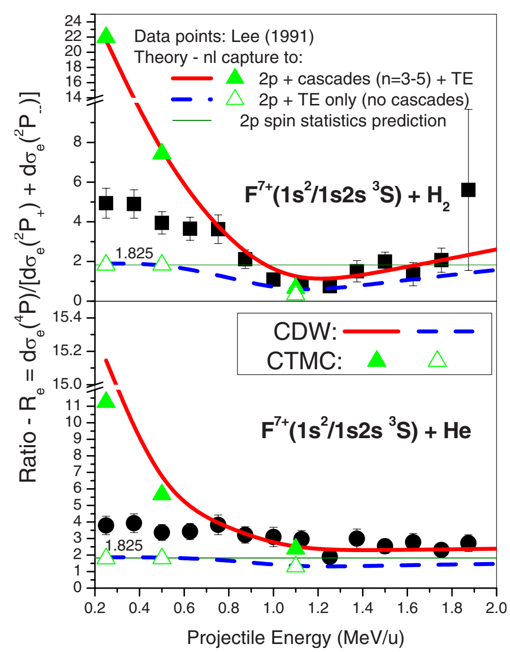

FIG. 5. (Color online) Ratio R_e of experimental and theoretical Auger SDCS shown in Fig. 4. The expected spin statistics ratio is 1.825 [28].

what extent dealignment effects at 0° observation [24,25] should also be considered given possible redistribution of magnetic substate populations due to feeding by cascades [26]. 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. For the H_2 target, the distinct hump in the doublet SDCS due to resonant transfer excitation (RTE) [5] is nicely reproduced by our calculations [3,27]. With decreasing collision energy below 0.7 MeV/u, feeding by cascades becomes increasingly more intense and even the $1s2s2p\ ^2P_{\pm}$ levels start to receive some cascade contributions. Our cascade analysis for $n \leq 5$ is found to seriously underestimate both quartet and doublet production at low collision energies for reasons not yet understood. The disagreement is seen to be worse for doublets produced in collisions with H_2 targets, seemingly ruling out nonresonant transfer excitation (NTE) as a possible cause, since this is known to be much weaker for H_2 than for He [27]. An upper limit CDW estimate on the Auger SDCS is also provided in Fig. 4 by assuming all capture to $n=2-8$ levels ends up directly into the $1s2s2p\ ^4P$ or $^2P_{\pm}$ states. 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 insufficient in the case of the ^4P levels, while for the $^2P_{\pm}$ levels, though more than sufficient, it seems not to arrive there.

The ratio of quartet to doublet SDCS directly from Fig. 4 is plotted in Fig. 5. R_e with both cascade and TE corrections (thick continuous red line) is clearly in better agreement with the data than without the cascade corrections (dashed 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 already observed disagreement in the absolute values of the computed SDCS is clearly evident.

In conclusion, we give a detailed analysis demonstrating the existence of a *selective* cascade-feeding mechanism resulting in the preferential population of low-lying long-lived metastable levels. For the particular collision system reported here, at collision energies above 0.7 MeV/u, this mechanism is found to be largely responsible for the recently reported nonstatistical enhancement of the $1s2s2p\ ^4P$ states [11]. Below 0.7 MeV/u, our understanding of both $^2P_{\pm}$ and ^4P production seems incomplete and further investigation is clearly necessary to understand the large discrepancies between theory and experiment. These results also underscore the significance of cascades, especially when metastable states are involved.

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