Electron Correlation in the Formation of Hollow States along the Li-like Isoelectronic Sequence

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Hollow-state (double K-shell vacancy) production in Li-like Be^+, B^{2+}, C^{3+}, and F^{6+} ions colliding with He has been investigated using high-resolution Auger-electron spectroscopy. The formation of discrete states and their relative intensities show striking dependences on the nuclear charge Z of the ion. From the projectile velocity dependences of these states the contribution of the electron-electron interaction is determined, showing that the hollow states are formed largely by electron correlation with a strength that diminishes for increasing Z.

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A fundamental area of interest in atomic physics is the study of dynamical electron correlation effects responsible for electronic transitions in collisions between atomic systems [1,2]. Correlation phenomena are manifested, for example, when electronic rearrangement accompanies electron promotion initiated by an incident photon or charged particle. In intermediate-to-high velocity collisions of charged particles with atoms where the perturbation picture is valid, multiple electronic transitions can be caused either by independent interactions with the charged perturber in a single collision event or by a single interaction with the perturber where accompanying transitions are due to correlation within the perturbed system. Hence, two categories of multiple electronic transitions are distinguished: (1) those resulting from independent first-order interactions between the perturber and the promoted electrons, and (2) those due to a single first-order perturber-electron interaction combined with one or more second-order electron-electron interactions. This formally more correct terminology is employed here instead of the two-step-two (TS2) and two-step-one (TS1) notations used previously for these cases, respectively [1]. In the latter case of TS1, excitation of a second electron is attributed to either a so-called *shake* [3] or a *dielectronic* [4] process.

Of particular interest is the study of electron correlation effects leading to hollow-state production in which doubly vacant K-shell states are formed in atoms and ions in the interaction with a perturber (photon or ion). Recently, attention has focused on hollow-state formation in atomic Li by incident photons [5,6], by incident ions [7,8], and most recently by incident electrons [9].

While the study of hollow-state formation in single collision events has been largely limited to atomic Li, there is widespread interest in studying hollow states along the Li-like isoelectronic sequence. For such systems, the effect that the changing Z plays in the role of correlation can be determined and the results used to provide sensitive tests for theoretical calculations and a detailed understanding of atomic structure. In general, as the charge of the parent Z becomes smaller, the relative strength of the electron-electron interaction increases and correlation effects are expected to become more prevalent. The present study provides the basis for future studies of Li-like ions excited in collisions with photons.

Here the formation of double K-shell vacancies in Li-like ions ($Z = 4$, 5, 6, 9) produced in collisions with neutral He is considered. It is well known that for Li-like species long-lived metastable states, which can complicate measured spectra, do not exist. The identification of hollow states is achieved with high-resolution measurements of Auger-electron emission from specific double K-shell vacancy intermediate excited states, and the role of electron correlation in their formation is determined from the collision velocity dependence. Not only does the existence of specific discrete hollow states depend on the parent Z, the intensities of these states also vary strongly with atomic number and with the collision velocity v_p.

The measurements were conducted at the Western Michigan University tandem Van de Graaff accelerator laboratory. After acceleration to energies of 0.5–2.0 MeV/u (velocities of 4–9 atomic units), the Li-like projectiles were directed into the scattering chamber where they collided with a neutral helium gas target. Singly and doubly K-shell excited Li-like ions were investigated by detecting Auger electrons emitted at 0° with respect to the beam direction using a high-resolution tandem parallel-plate electron spectrometer.

Auger spectra for electron emission from double K-shell vacancy states for the Li-like ions investigated are shown in Fig. 1 (the indicated charge states are those of the incident ions). For ions other than Be^+ the spectra contain contributions from singly K-shell excited states intermixed with the doubly K-shell excited states (hollow states are patterned to distinguish them from single K-shell excitation lines). Absolute cross sections for Auger emission were determined by normalizing the measured yields to auxiliary measurements for elastically scattered (binary encounter) electrons [10,11].

From Fig. 1, it is clear that the changing Z of the parent ion has a strong influence over which hollow states are

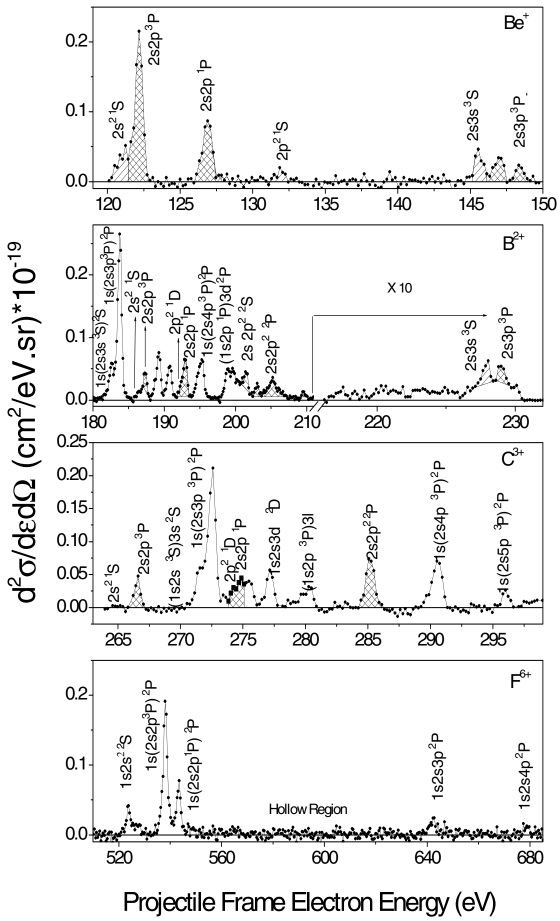

FIG. 1. Measured Auger spectra for hollow-state formation in collisions of Be^+, B^{2+}, C^{3+}, and F^{6+} like ions with He at 1 MeV/u. Hollow states are distinguished by the patterned regions under the peaks: diagonal-patterned areas, S states; crosshatched areas, P states.

produced, as well as the relative intensities of the lines. The spectrum for Be^+ ions looks very similar to the corresponding spectrum for Auger emission from Li following collisions with fast charged particles [7,9] and photons [5], for which the dominant hollow states are produced via K-shell ionization plus excitation to form $2l2l'$ and $2l3l'$ configurations. In the case of B^{2+} ions, states with these same two-electron hollow configurations are observed, as well as three-electron hollow states with configurations $2l2l'2l''$ corresponding to double K-shell excitation. For C^{3+} ions hollow $2l2l'$ states (ionization plus excitation) and hollow $2l2l'2l''$ states (double excitation) are formed, but no $2l3l'$ states are present. For F^{6+}, the cross section for hollow-state production was too small to be measured. The hollow-state configurations observed at different collision velocities for each of the ions studied are summarized in Table I.

Within the framework of perturbation theory, it has been well documented that the contribution of correlation can be determined by measuring the yield of doubly vacant states

TABLE I. Configurations observed (indicated by "X") in measurements of Auger emission from incident Li-like ions following collisions with He at velocities between 4–9 a.u. (~0.5–2.0 MeV/u) (see Fig. 1).

	$2l2l'$	$2l3l'$	$2l2l'2l''$
Be^+	X	X	...
B^{2+}	X	X	X
C^{3+}	X	...	X
F^{6+}	...	...	...

as a function of the collision velocity [1]. Specifically, for collisions of charged particles within the Born approximation ($Z/\nu_p < 1/2$) single excitation and ionization have the form $Z^2[\ln\nu_p]^m/\nu_p^2$, with $m = 0, 1$ for monopole ($1s \rightarrow ns,\ \varepsilon s$) and dipole ($1s \rightarrow np,\ \varepsilon p$) transitions, respectively. Cross sections for single K-shell excitation, i.e., $1s \rightarrow nl$, obtained in the present work (not shown) were found to be in excellent agreement with this prediction [12,13] for $Z_{\rm eff} = 2$, attesting to the validity of the Born approximation. The fact that the perturbing He atom has an effective charge $Z_{\rm eff} = 2$ indicates that at these small impact parameter collisions the screening of the He nucleus by its electrons, as well as projectile-target electron-electron interactions, are negligible [14]. Furthermore, the cross section for double K-shell vacancy production caused by independent first-order interactions between the projectile and the target electrons is expected to have the form ν_p^{-4}, while the cross sections for states due to a combination of first-order and second-order interactions should behave as $[\ln\nu_p]^m/\nu_p^2$ [1,3]. This latter dependence is the same as for single K-shell vacancy production because a single nucleus-electron interaction initiates electron promotion with the electron-electron interaction causing additional excitation. For states formed both by independent first-order interactions and by a combination of first- and second-order processes, the coherent sum of amplitudes for these processes determines the behavior of the cross section and may lead to interference effects.

The measured and calculated velocity dependences for formation of the $2s^2\,^1S$ and $2s2p\,^3P$ states for Be^+, B^{2+}, and C^{3+} ions are shown in Fig. 2. The solid curves show the expected behavior for double K-shell vacancy formation of these states resulting from the combination of single first-order and second-order interactions ($[\ln\nu_p]/\nu_p^2$ because ionization is dominated by $1s \rightarrow \varepsilon p$ dipole transitions), while the dashed curves (shown only for Be^+) give the predicted behavior for independent first-order interactions between the promoted electrons and the perturbing nucleus (ν_p^{-4}). From these velocity dependences it is concluded that the $2s^2\,^1S$ and $2s2p\,^3P$ states are populated in large part by second-order interactions (electron correlation), particularly at the highest collision velocities investigated

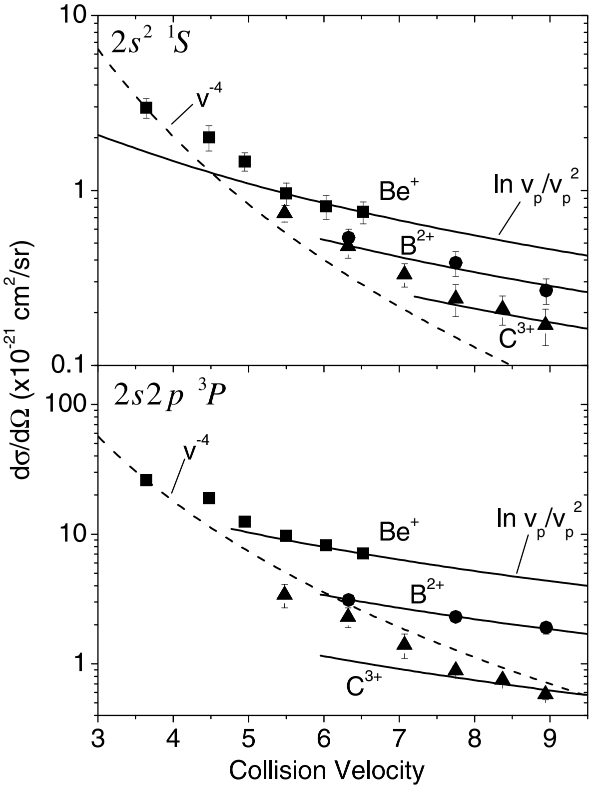

FIG. 2. Measured cross sections for formation of the $2s^2\,{}^1S$ (upper panel) and $2s2p\,{}^3P$ (lower panel) hollow states for incident Be^+, B^{2+}, and C^{3+} ions as a function of collision velocity (in atomic units). The solid curves represent the $[\ln\nu_p]/\nu_p^2$ behavior expected for two-electron transitions caused by the combination of first-order and second-order interactions. The dashed curves show the ν_p^{-4} dependence associated with independent first-order interactions leading to two-electron transitions.

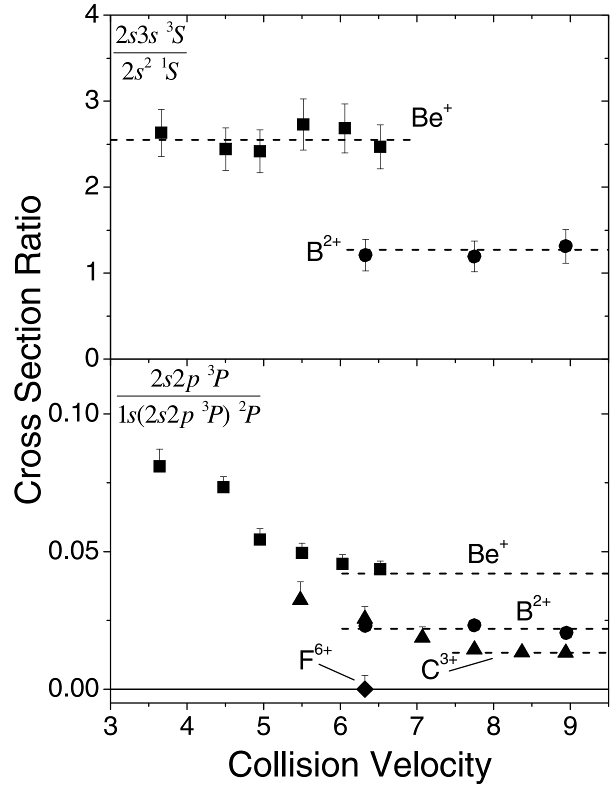

FIG. 3. Upper panel: Ratio of the $2s3s\,{}^3S$ (hollow) state to the $2s^2\,{}^1S$ (hollow) state for incident Be^+ and B^{2+} ions as a function of collision velocity (in atomic units). The dashed lines are constant values of this ratio drawn to guide the eye. Lower panel: Ratio of the $2s2p\,{}^3P$ (hollow) state to the $1s(2s2p\,{}^3P)\,{}^2P$ (single K-shell vacancy) state for incident Be^+, B^{2+}, C^{3+}, and F^{6+} ions as a function of collision velocity. For F^{6+} the result is plotted at zero with an upper bound determined from the signal-to-noise ratio and the resolution of the measurement.

for each ion. Also evident is the strong dependence of the cross section on the atomic number of the parent ion.

In order to determine the effect of the parent Z on the electron correlation strength, we consider the cross-section ratios $2s3s\,{}^3S/2s^2\,{}^1S$ and $2s2p\,{}^3P/1s(2s2p\,{}^3P)\,{}^2P$ shown in Fig. 3 (the $2s3s\,{}^3S$ state was observed only for Be^+ and B^{2+}). The $2s3s\,{}^3S/2s^2\,{}^1S$ ratio (upper panel) is essentially independent of velocity, indicating that the relative population of the two states is caused mainly by the electron-electron interaction. Furthermore, the measured ratio for Be^+ is about twice that of B^{2+}, and the $2s3s\,{}^3S$ state was not observed for higher Z, showing that the Z of parent ion strongly influences the formation of these states (via correlation) with $2s3s\,{}^3S$ formation being rapidly damped.

As done earlier for atomic Li [8], the $2s3s\,{}^3S/2s^2\,{}^1S$ ratios can be compared with a two-step shake (TS1) model [3] for the production of these S states, where an angular momentum conserving monopole transition, $1s \to ns$, follows an initial K-shell ionizing event. The probability for transitions due to shake is determined by projecting the wave function for a $1s$ electron in the ground state of the three-electron system onto the eigenstates of the excited two-electron ion. In this manner, shake probabilities were calculated from the overlap of the initial "frozen" two-electron $1s_0 2s_0\,{}^3S$ state (following ionization) with the $2s3s\,{}^3S$ state of the excited He-like ion, and from the overlap of the initial frozen $1s_0 2s_0\,{}^1S$ state with the excited $2s^2\,{}^1S$ state, using the multiconfiguration Hartree-Fock (MCHF) code of Fischer [15].

The results of these calculations are shown in Table II, where it is seen that the shake probabilities decrease strongly with the parent Z. For the $2s3s\,{}^3S$ state there are two pathways from the $1s_0 2s_0\,{}^3S$ state: a direct ($1s_0 \to 3s$) transition or a two-step ($1s_0 \to 2s + 2s_0 \to 3s$) exchange process. Because these pathways lead to the same final state, there can be interference between them. From Table II the exchange term is seen to be larger than the direct term for Be^{2+} and B^{3+} ions (also for Li^+), indicating that the two transition exchange process is more probable

TABLE II. Shake probabilities for the formation of the $2s^2\,{}^1S$ and $2s^2\,{}^1S$ states in Li^+, Be^{2+}, B^{3+}, and C^{4+} from initial "frozen" $1s_0 2s_0\,{}^{1,3}S$ states. The last column shows the expected $2s3s\,{}^3S/2s^2\,{}^1S$ ratio based on the calculated probabilities.

Ion	$\vert\langle 2s^2\,{}^1S\vert 1s_0 2s_0\,{}^1S\rangle\vert^2$	$\vert\langle 2s3s\,{}^3S\vert 1s_0 2s_0\,{}^3S\rangle\vert^2$ Direct	Exchange	Interference	Total	Ratio: ${}^3S/{}^1S$
Li^+	0.0081	0.0005	0.0090	0.0042	0.0140	1.7
Be^{2+}	0.0059	0.0010	0.0030	0.0033	0.0073	1.2
B^{3+}	0.0043	0.0008	0.0011	0.0018	0.0037	0.9
C^{4+}	0.0031	0.0006	0.0005	0.0011	0.0022	0.7

than a single direct transition. The interference term also contributes significantly to the formation probability for $2s3s\,{}^3S$ and with increasing Z becomes larger relative to the exchange. For both Be^{2+} and B^{3+} the calculated ${}^3S/{}^1S$ shake ratios underestimate the measured ratios (see Fig. 3), a result that is consistent with that found for Li^+ [8]. This underestimation of the measured ratios likely points to the inadequacy of the shake model, and suggests that electron correlation effects cannot be intrinsically isolated from the initial electron promotion event (by a photon or a charged particle) as is done in the typical two-step formulation (TS1) of correlation.

The ratios of the hollow (double K-shell vacancy) $2s2p\,{}^3P$ state to the single K-shell excitation $1s(2s2p\,{}^3P)\,{}^2P$ state (lower panel of Fig. 3) are similar to measurements of the ratio of double to single K-shell ionization of helium [1]. However, a significant distinction is that the present ratios involve discrete excited states for which shake cannot produce the $2s2p\,{}^3P$ state from the $1s_0 2s_0\,{}^3S$ state because shake must conserve angular momentum. From the figure, the $2s2p\,{}^3P/1s(2s2p\,{}^3P)\,{}^2P$ ratios are seen to approach constant values for the highest collision velocities investigated. As for He, a constant asymptotic limit indicates that electron correlation is the dominant mechanism for double vacancy production [1], since only a single first-order interaction is responsible for single and double vacancy production. Thus, these asymptotic limits give the relative electron correlation strengths for the different ions investigated, showing that the strength decreases significantly as the Z of the parent ion increases.

In summary, we have investigated the formation of discrete hollow states produced in incident Li-like ions colliding with He atoms. From the velocity dependences of the observed hollow states, electron correlation is shown to contribute substantially to their production. The presence of particular doubly vacant K-shell states and their intensities vary strongly with the atomic number of the parent ion, pointing to significant variations in the correlation strength. These results should be useful for detailed testing of theoretical calculations of electron correlation strengths along the Li-like isoelectronic sequence. The present work lays the foundation for direct correlation studies involving interactions between ions and single photons.

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