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Identification and characterization of the dielectronic process in the formation of two *K*-shell vacancies in atomic Li by fast electron impact

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Electron correlation effects in the production of two *K*-shell vacancies in lithium are investigated by bombarding atomic Li with 5-keV electrons. High-resolution Auger electron spectra show that discrete doubly vacant *K*-shell states are predominantly formed from ionization plus excitation, giving rise to the $2sns\,^{1,3}S$ and $2snp\,^{1,3}P$ states of Li^+. For these fast electron projectiles, the shake-up and dielectronic manifestations of the electron-electron interaction are shown to be the only mechanisms for creating the observed hollow states. The formation of the $2snp\,^{1,3}P$ states is found to be mediated only by the dielectronic process, permitting this aspect of the electron-electron interaction to be separately identified and its contribution characterized.

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While the most basic three-body processes such as electron-impact single ionization of atomic hydrogen are now essentially understood [1], the investigation of many-electron systems remains the subject of intensive research activities. In this latter regard, extensive efforts continue to be devoted to the identification of electron correlation phenomena associated with electronic transitions induced by photon or charged particle impact on atomic targets [2]. For instance, with the help of improved experimental and theoretical techniques, new attention has focused on two-electron processes in helium [3–10], the simplest atom containing more than one electron.

The understanding of many-electron processes is often formulated by isolating mechanisms that manifest themselves in characteristic features of the measured or calculated observables. A prime example is double ionization of He. For photon impact, double ionization is generally described in terms of two mechanisms, shake-off and a dielectronic process [2]. Similar mechanisms are invoked in the case of fast charged particle impact [2,11,12] where, additionally, both electrons can be removed in independent interactions with the projectile. In the shake-off approach it is assumed that one of the target electrons is removed suddenly by the impacting photon or particle, while the other electron remains initially unperturbed. Then, because of the sudden change in the atomic Coulomb field, this latter electron relaxes from its initial He wave function onto the hydrogenic set of He^+ wave functions, which includes continuum states, leading to a probability for the second electron to be shaken off (ionized). On the other hand, the dielectronic mechanism describes the correlated dynamics of the two electrons as they leave the atom, where the primary electron ejects the secondary electron in a binary encounter. This final-state correlation process involves the mutual scattering of the electrons and is dielectronic in nature [11]. (This dielectronic mechanism has alternatively been referred to as TS (two-step process with one projectile interaction) [2], ‘‘knockout’’ [10] or ‘‘kickout’’ [13].) Similar shake (shake-off or shake-up) and dielectronic processes have been invoked to describe double excitation [14] and ionization-excitation transitions [15].

Apart from general characteristics such as the prevalence of shake when the primary electron is ejected with high kinetic energy, the separation of the shake and dielectronic processes and the determination of specific properties associated with each remains a challenging task, both experimentally and theoretically. The first experimental identification of the shake and dielectronic processes was made for doubly vacant *K*-shell states in lithium resulting from fast ion impact [16] and more recently for the double ionization of helium induced by incident photons [10]. In the case of fast ions, state-selective measurements were used to separately identify the dielectronic and shake processes based on the fact that they primarily contribute to the formation of different excited states [16]. However, as noted above, for fast ions independent interactions with the projectile (sometimes referred to as TS2) can also give rise to the observed states, making it difficult to isolate the electron-electron processes. For the double ionization of helium by photon impact, the energy sharing and angular asymmetry of the ejected electrons were used to determine characteristics that are generally associated with shake and dielectronic processes, respectively [10]. In this case, both electrons are ejected to the continuum, representing a summation over all final states, so the observed properties of the ejected electrons involve a continuous progression from shake to dielectronic processes that are difficult to isolate. The characteristics of the electron-electron interaction processes were determined for a limited number of electron energies for the primary ejected electron [10]. Hence, the measured contribution to double ionization for the events that were found to involve mainly dielectronic processes constituted a small fraction of the total of such events.

In the present work, electron correlation effects for doubly vacant *K*-shell states resulting from ionization-excitation are investigated. By considering electron correlation involving

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discrete states, it will be shown that the production of specific excited states can be exclusively attributed to the dielectronic process. Specifically, the formation of two *K*-shell vacancies in lithium is investigated by bombarding atomic Li with 5-keV electrons. For atomic Li, electron correlation effects associated with *K*-shell ionization-excitation events can be readily studied from the resulting Auger-electron emission (this is not possible for He). Furthermore, by using fast electrons as projectiles, two-electron transitions due to independent interactions with the projectile [two-step process with two projectile interactions (TS2)] will be shown to be negligible. Thus, in contrast to previous work with fast Ar^{18+} ions [16], for the present investigation the shake and dielectronic processes are the only mechanisms by which double *K*-shell vacancies are created in the target atom. Under these conditions, we show that a "pure" dielectronic contribution to the formation of hollow lithium can be isolated experimentally.

The present measurements were conducted at CIRIL in Caen using an electron gun of simple design. A beam of 5-keV electrons of intensity $\sim 100\ \mu\mathrm{A}$, collimated to a diameter of ~ 2 mm was directed onto a jet of lithium vapor atoms. The Li target was obtained by heating metallic Li in a temperature-controlled oven. The lithium temperature was ~ 700 K to obtain a stable jet of Li atoms without producing significant amounts of molecular Li_2 [15]. Electron emission from the Li target was investigated using an electrostatic parallel-plate electron spectrometer [17]. The voltage on the plates of the spectrometer was scanned to record the electron yields as a function of the electron emission energy. To obtain the angular dependence of electron emission, spectra were collected at several observation angles relative to the beam direction.

Figure 1 shows Auger spectra recorded at various electron emission angles for single *K*-shell excitation. The peaks originate from the Auger decay of doubly excited states $1s2lnl'\,^2L$ $(n \geqslant 2)$ of Li. Small contributions from molecular Li_2 are observed near 52 eV. However, the total Li_2 contribution does not exceed 5% of the total measured electron yield. The spectra are dominated by the strong $1s(2s2p\,^3P)\,^2P$ line formed by the $1s \rightarrow 2p$ dipole transition from the ground-state $1s^2 2s\,^2S$. The angular dependence of this line intensity was verified to be symmetric with respect to the emission angle $\theta = 90°$. A $\sin^2\theta$ dependence that reproduces the maximum at 90° was found, indicating that the magnetic quantum number $M_L = 1$ is predominantly populated [19]. In addition to the $1s(2s2p\,^3P)\,^2P$ line, significant line intensities are observed for other $1s2snp\,^2P$ states, resulting from dipole transitions $1s \rightarrow np$ involving higher n values $(n > 2)$. On the contrary, the line intensity for the $1s2s^2\,^2S$ state (near 51 eV) populated via the monopole transition $1s \rightarrow 2s$ is found to contribute negligibly to the spectra. Similar results were previously obtained for 95-MeV/u Ar^{18+} projectiles [16].

From the spectra of Fig. 1, it is found that the intensity of the main $1s \rightarrow 2p$ dipole transition [giving rise to $1s(2s2p\,^3P)\,^2P$] is about 30 times larger than that for the $1s \rightarrow 2s$ monopole transition (leading to $1s2s^2\,^2S$). Since single excitation results from a direct interaction between the projectile and the active target electron, i.e., a projectile-electron $(p\text{-}e)$ interaction, the observation that single excitation produces mainly P states indicates that for the present collision system transitions mediated by the $p\text{-}e$ interaction are predominantly dipole. This result is important in determining the mechanisms for producing two *K*-shell vacancies in lithium, for which it will also be assumed that transitions due to $p\text{-}e$ interactions are mostly dipole.

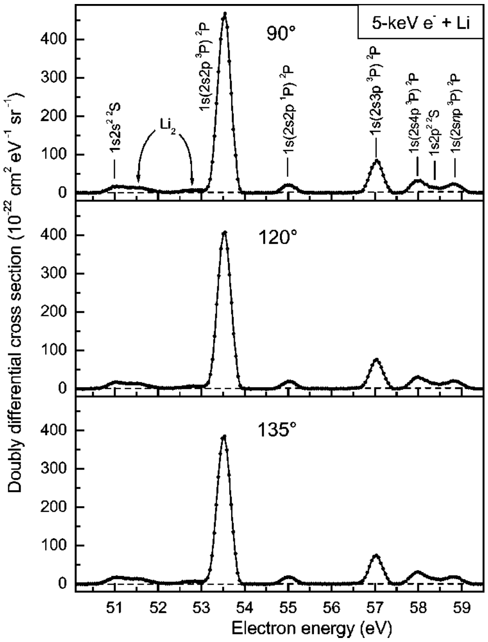

FIG. 1. Single-*K*-shell excitation Li Auger spectra for electron emission angles of 90°, 120°, and 135° induced by 5-keV electron impact. The specific excited-state configurations are indicated. The background due to continuous electron emission from direct ionization of the target has been subtracted. Absolute cross sections were determined by normalizing the large $1s(2s2p\,^3P)\,^2P$ line to the theoretical cross section obtained in the plane-wave Born approximation [18].

High-resolution electron spectra for double-*K*-shell vacancy production are shown in Fig. 2. The main features of these spectra are shown to be nearly independent of the observation angle. Most of the observed peak intensity originates from the Auger decay of doubly *K*-shell-excited states of Li^+, indicating that for fast electron impact *K*-shell ionization plus excitation events are significantly more probable than double-*K*-shell excitation events. This was also the case for fast ion [16] and photon [20] impact. The main lines are attributed to the $2sns\,^{1,3}S$ and $2snp\,^{1,3}P$ states of Li^+, while the only evidence for double-*K*-shell excitation is the $2s^2 2p\,^2P$ line centered at about 75.5 eV. Double-*K*-shell ionization cannot be separately identified with the method used here because no Auger emission will result.

The important feature of these spectra is the fact that, contrary to the case of single-*K*-shell excitation, the line intensities for the S and P states corresponding to double-*K*-shell vacancies are of the same order of magnitude. Hence, both monopole and dipole transitions play comparable roles in the formation of two *K*-shell vacancies, suggesting a different mechanism for the formation of these double-*K*-shell vacancy states compared to single-*K*-shell vacancy states.

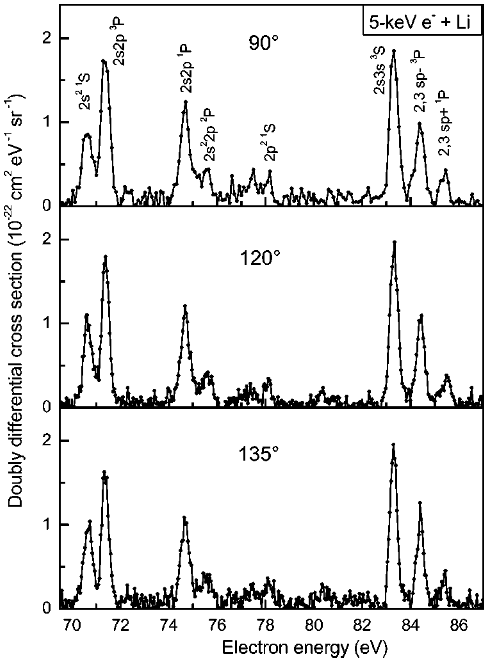

FIG. 2. Double-*K*-shell vacancy Li Auger spectra for electron emission angles of 90°, 120°, and 135° induced by 5-keV electron impact. The excited-state configurations are indicated. The background due to continuous electron emission from direct ionization of the target has been subtracted. Absolute cross sections were determined as in Fig. 1.

Additionally, the second *K*-shell vacancy results solely from the electron-electron (*e*-*e*) interaction. This can be deduced from Table I where the relative intensities for the double-*K*-shell vacancy states produced by electron impact are seen to be nearly the same as those for photon impact from Ref. [20]. For incident photons, two *K*-shell vacancy states can only result from the *e*-*e* interaction following an initial *K*-shell ionization (or excitation) event caused by the incident photon, which is assumed to interact with a single electron. The fact that the relative intensities of the double-*K*-shell vacancy states are nearly the same for incident photons and the incident fast electrons considered here implies that the mechanisms for the production of these states are the same.

For these fast electron-atom collisions, independent projectile-electron interactions, i.e., TS2, could also give rise to the formation of the $2snp\ ^{1,3}P$ states via the dipole transitions $1s \rightarrow \varepsilon p$ and $1s \rightarrow np$. If this were the case, however, then the observed relative line intensities for these *P* states would be larger than those observed for incident photons. Such was the case for fast heavy ion impact as seen from the results for 95 MeV/u Ar^{18+} ions that are also listed in Table I, where the relative intensities for the $2snp\ ^{1,3}P$ states are clearly larger than those for electron or photon impact.

Thus, it is concluded that the second *K*-shell vacancy for 5-keV electron impact results almost entirely from the *e*-*e* interaction following ionization (or excitation) of the first target *K*-shell electron by the incident fast electron. This result is consistent with plane-wave Born approximation (PWBA) calculations (not shown) [18] that indicate the TS2 contribution to be negligible when using projectiles for which the perturbation strength $|Z|/v$ (Z is the projectile charge and v is the collision velocity) is smaller than 0.1 a.u., as is the case here.

TABLE I. Relative intensities, normalized to the $2s^2\ ^1S$ line, of the Li^+ double-*K*-shell vacancy states observed in the present work for 5-keV electrons compared with those of Ref. [20] for 197-eV photons and those of Ref. [16] for 95-MeV/u Ar^{18+} ions. For this work, the relative intensities were obtained from total cross sections that were determined by integrating the high-resolution Auger spectra (Fig. 2) over electron emission energy and angle. The relative intensities for Ref. [20] were determined from Fig. 2 of that work. The uncertainties are 10–15 % for the relative intensities of this work and Ref. [20], while the uncertainties for the relative intensities of Ref. [16] are 15–20 %.

	Relative intensities		
States	Present work 5-keV e^-	Ref. [20] 197-eV photons	Ref. [16] 95-MeV/u Ar^{18+}
$2s^2\ ^1S$	1	1	1
$2s2p\ ^3P$	1.4	1.1	2.6
$2s2p\ ^1P$	1.1	0.8	1.3
$2p^2\ ^1S$	0.2	0.2	0.4
$2s3s\ ^3S$	1.3	1.6	1.7
$(2,3\,sp\text{-})\ ^3P$	0.9	1.2	1.5

Since the formation of doubly vacant *K*-shell states in lithium by 5-keV electron impact can be considered to be entirely governed by the *e*-*e* interaction, only the shake and dielectronic processes need to be considered in giving rise to these states. For both mechanisms, the primary interaction of the projectile with the target produces an intermediate state that becomes the initial state for the subsequent *e*-*e* process. From the present single-*K*-shell excitation results, the primary transition is principally dipole, i.e., $1s \rightarrow \varepsilon p$ for ionization or $1s \rightarrow np$ for excitation. On the other hand, the subsequent transition mediated by the *e*-*e* interaction can be either monopole or dipole, as evidenced by the fact that the *S* and *P* states associated with two *K*-shell vacancies are comparable in magnitude (see Fig. 2).

As mentioned above, in the case of the shake mechanism the primary transition is assumed to take place suddenly, in a time significantly shorter than the characteristic periods of the target electrons. The primary *K*-shell process is "sudden" if one of the 1*s* electrons is ejected with high kinetic energy. Then, the remaining target electrons experience the relaxation of the electron cloud, while the ionized electron is already far removed from the target. In this picture, the residual target ion is isolated when the shake process takes place. Hence, the total angular-momentum L of the transient target ion cannot change, and the shake process can only give rise to $\Delta L = 0$ (and $\Delta l = 0$) transitions [16]. Since the primary *K*-shell ionization creates a transient state $1s2s\ ^{1,3}S$ of Li^+, the subsequent *K*-shell excitation via shakeup can only take place by a $1s \rightarrow ns$ monopole transition, leading to the $2sns\ ^{1,3}S$ states. Consequently, the shake-up process cannot contribute to the formation of the $2snp\ ^{1,3}P$ states for which a $\Delta L = 1$ (and $\Delta l = 1$) transition is required.

In contrast to the shake-up process, the dielectronic process can involve an exchange of angular momentum between the primary and secondary active electrons due to the mutual ‘‘scattering’’ of these electrons. For instance, when the primary ionization occurs by means of a $1s \rightarrow \varepsilon p$ transition, the dielectronic process can cause excitation by a subsequent dipole transition $1s \rightarrow np$ leading to two K-shell vacancies if the εp electron, as it leaves the atom, promotes the remaining $1s$ electron to np by giving up its $l=1$ unit of angular momentum [16]. While the dielectronic process does not necessarily have to involve an exchange of angular momentum, thereby also permitting monopole transitions that give rise to the $2sns\ ^{1,3}S$ states of Li^+, only the dielectronic manifestation of the e-e interaction can produce the observed $2snp\ ^{1,3}P$ states. Since these latter states are also not formed from independent interactions with the projectile (TS2) in the present electron-lithium collisions (see Table I), as discussed above, the dielectronic process is the only mechanism for producing the observed $2snp\ ^{1,3}P$ states. Thus, spectral identification of the double-K-shell vacancy P states in the present work reveals directly the dielectronic aspect of the e-e interaction, from which its specific role can be ascertained.

From the relative intensities listed in Table I, the $2snp\ ^{1,3}P$ states for 5-keV electron impact constitute about 60% of the total double-K-shell vacancy intensity associated with K-shell ionization plus excitation events (Fig. 2). Moreover, the dielectronic process may contribute (in addition to shake-up) to the formation of the $2sns\ ^{1,3}S$ states in Li^+, although this contribution is expected to be small. Hence, the dielectronic process, which is the only mechanism for producing the P states, plays the dominant role in the production of hollow Li^+ ions by fast electron impact. In the case of 95-MeV/u Ar^{18+} ion impact [16], these same P states also constitute the largest contribution to the formation of the double-K-shell vacancy states. However, a considerable fraction (more than half as can be deduced from Table I) of this contribution arises from independent projectile-electron interactions. On the other hand, when using projectiles for which the perturbation strength $|Z|/v$ is smaller than 0.1 a.u., as is the case here with fast electron projectiles, the dielectronic contribution to double-K-shell vacancy state formation is separately identified and its specific contribution determined. Furthermore, compared to the case of double-ionization events [10] where both electrons are ejected to the continuum, the present results for ionization plus excitation events exhibit the advantage of investigating *discrete* excited states in order to determine specific characteristics associated with dielectronic and shake processes, respectively.

In summary, we have investigated electron correlation effects in the production of doubly vacant K-shell states in atomic lithium induced by 5-keV electrons. Spectra obtained by means of high-resolution Auger spectroscopy show that two K-shell vacancies involving discrete states are produced mainly by K-shell ionization plus excitation events, leading to $2lnl'$ $(n \geqslant 2)$ configured S and P states of hollow Li^+. For these high-velocity electron projectiles, independent interactions with two target electrons (TS2) have been shown to play a negligible role so that only processes associated with the e-e interaction (dielectronic or shake-up) lead to the observed double-K-shell vacancy states. Since the shake-up process cannot create the $2snp\ ^{1,3}P$ states from the intermediate $1s2s\ ^{1,3}S$ states, the formation of these P states is essentially mediated only by the dielectronic process. Thus, the dielectronic manifestation of the e-e interaction can be separated from shake and its contribution quantitatively determined for these fast charged particle-atom collisions. Based on the present results, the dielectronic process, i.e., dynamic electron correlation, is found to be responsible for at least half of the e-e interaction events that lead to discrete double-K-shell vacancy states in 5-keV electron-lithium collisions. Future work will focus on the formation of the $2sns\ ^{1,3}S$ states, which can generally involve both shake and dielectronic processes, to determine the relative importance of these e-e mechanisms in producing the S states.

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