Production of Hollow Lithium by Multielectron Correlation in 95 MeV/nucleon Ar^{18+} + Li Collisions

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Double-K-shell-vacancy production in Li by 95 MeV/nucleon Ar^{18+} projectiles was investigated using high-resolution electron spectroscopy. The two K vacancies are found to come about mainly by ionization and excitation, giving rise to excited states in hollow Li^+. Strong line intensities from the $2s^2\,{}^1S$ and $2s3s\,{}^3S$ configurations provide spectral identification for the electron-electron interaction. Production of the $2s3s\,{}^3S$ state, with an intensity greater than that for $2s^2\,{}^1S$, is attributed to a three-electron transition involving correlation.

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Multielectron transitions in atoms provide insight into the fundamental nature of atomic structure by probing dynamical electron correlation effects. In recent years, the importance of the electron-electron (e-e) interaction in understanding the multiple excitation or ionization of atoms has been widely recognized [1–6].

In ion-atom collisions, multielectron transitions can result from the e-e interaction or from the nucleus-electron (n-e) interaction (n here not to be confused with neutron). If a multielectron transition leads to an empty K shell, then a so-called "hollow atom" (or ion) is produced. Such double-K-shell vacancy production in a target atom by ion impact can be caused by separate n-e interactions, or by an n-e interaction followed by an e-e interaction. In the former case, the process is referred to as TS2 (two-step with two projectile interactions), and in the latter case it is called TS1 (two-step with one projectile interaction), a process which implies dynamic electron correlation [4,6]. Such dynamic correlation is well known in photoionization [2,7], where multielectron transitions resulting from a single photon impact can be caused only by the e-e interaction. Thus, hollow-atom production by photon impact corresponds to TS1 in the case of ion impact. During the past decade, double-K-shell ionization of He by fast ions and photons has attracted much interest [8] due to the insight it provides into dynamic correlation effects.

Dynamic electron correlation generally falls into two categories, corresponding to whether the first electron is emitted slowly or suddenly. For slow emission, subsequent excitation or ionization of a second electron involves the mutual scattering of two electrons, i.e., it is *dielectronic* in nature [4]. On the other hand, sudden emission can result in a subsequent electron transition due to the change in potential seen by the second active electron as the excited system relaxes [2]. This latter type of transition is a "mean-field" effect referred to as a *shake* process.

To investigate dynamic correlation in atomic collisions, we consider the formation of hollow Li caused by fast projectiles. Early observations of hollow Li with incident ions of intermediate energy were made by Ziem *et al.* [9] and Rødbro *et al.* [10]. More recently, hollow Li has been investigated using photoionization [11–14] and fast ions [15]. In addition to these works, Müller *et al.* [16] have investigated hollow Li production in collisions of Li^+ ions with electrons.

Double-K-shell vacancy (hollow lithium) production is shown schematically in Fig. 1 for ground-state Li ($1s^2 2s$). In the figure, ionization via a $1s \rightarrow \varepsilon p$ dipole transition is associated with excitation via a $1s \rightarrow 2l$ transition (for $l = 1$, the excitation is dipole, while for $l = 0$ it is monopole). For fast ions, dipole transitions are expected to come about mainly by n-e interactions [17]. On the other hand, monopole transitions can be induced by either the n-e interaction or the e-e interaction. Furthermore, shake processes, which are a manifestation of the e-e interaction, can give rise to *only* $\Delta l = 0$ transitions because these processes are an internal rearrangement of the residual ion, with the consequence that the total orbital angular momentum of the system cannot change.

It is important to note that ionization must occur first, as shown in Fig. 1, to give rise to the indicated double-K-shell-vacancy S states. Specifically, single-K-shell excitation occurring first would produce mainly the $1s(2s2p\,{}^{3,1}P)\,{}^2P$ or $1s(2s3p\,{}^3P)\,{}^2P$ configurations via n-e interactions, following which subsequent K-shell ionization cannot give rise to the $2s^2\,{}^1S$ and $2s3s\,{}^3S$ configurations. Hence, a definite time ordering [18,19] is

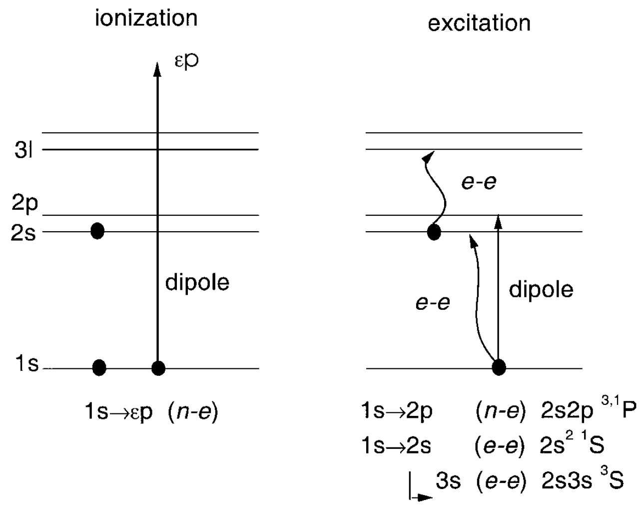

FIG. 1. Schematic showing double-K-shell-vacancy production in Li. Ionization is assumed to occur first, followed by excitation. The most prominent transitions are shown, as well as the dominant interaction involved (n-e or e-e) in producing the indicated double-K-shell-excited intermediate states.

required to produce the indicated double-K-shell vacancy S states. Since the probability for monopole transitions via the n-e interaction is small, the S states are expected to be produced mainly by the e-e interaction, and, consequently, dielectronic and shake processes require time ordering.

In view of these ideas, we see that identification of specific spectral features associated with double-K-vacancy production can provide insight into the importance of the e-e interaction in ion-atom collisions. A high-resolution Auger emission spectrum for hollow lithium was recently reported in the photoexcitation of Li [13], where the double-K-shell-vacancy states can result from only the e-e interaction.

In this work we investigated double-K-vacancy production in 95 MeV/nucleon ($v/c = 0.42$) Ar^{18+} + Li collisions. At such velocities, excitation or ionization by ions is expected to closely resemble that of a photon [17]. High-resolution measurements of Auger electron emission show that the two K vacancies are produced mainly by ionization and excitation events. (We cannot observe double-K-shell ionization in this work.) Observation of specific excited-state configurations makes possible the spectral identification of electron correlation and, additionally, provides strong evidence for a correlated three-electron transition.

The present measurements were carried out at the GANIL accelerator facility in Caen, France, using techniques similar to those used previously [20]. Briefly, the Ar^{18+} beam, of intensity 1–2 μA, was incident on a Li vapor jet target produced by resistive heating of metallic Li in a small oven. The beam was collimated to a size of about 2 mm $\times$ 2 mm, and the Li vapor target had a diameter of about 4 mm. Electrons (continuum and Auger) emitted from the Li target were observed with a parallel-plate spectrometer. Details of the scattering chamber and the electron spectrometer have been described previously [21].

The Li target posed several experimental challenges. First, the metallic lithium had to be heated slowly to drive contaminants from the surface. Then, the lithium temperature was set just high enough above the melting point (180 °C) to obtain a stable jet of Li atoms without producing significant amounts of molecular lithium, i.e., Li_2. For the temperatures used and the oven geometry, it is estimated that resulting target thickness was about 1×10^{14} atoms/cm^2. Furthermore, background electrons accelerated by the electric field associated with the relatively large current used to heat the metallic lithium had to be guarded against. During the measurements, the spectra were monitored continuously for accuracy.

In Fig. 2 are shown the measured spectra, differential in energy, for angles of 90°, 120°, and 160° relative to the beam direction (spectra were also measured for 60° and 140°). The background due to continuous electron emission resulting from ionization of the target, typically of the same order of magnitude as the observed peak intensities in the double-K-shell vacancy region, has been subtracted. The prominent single K-shell vacancy lines between 50 and 60 eV are indicated, as well as the main double-K-shell-vacancy lines between 70 and 85 eV. A small Li_2 contamination is observed between about 51.5 and 52.5 eV. These high-resolution spectra were obtained by decelerating the electrons to an energy of 10 eV prior to entering the parallel-plate analyzer. The resulting constant instrumental energy resolution ($\sim$0.4 eV) was considerably larger than the natural linewidths (<0.1 eV), so that the observed peak widths were also constant. The spectra were fit by fixing the peak positions to known energy values [9,10] and the peak widths to the experimental value. Absolute total cross sections, listed in Table I, were determined by normalizing the large $1s(2s2p\,^3P)\ ^2P$ single-K-excitation line to earlier data for this same collision system [22], and integrating over angle. The agreement with Born theory [23] is seen to be excellent.

The single-K-shell vacancy lines in Fig. 2 are all well-known configurations in neutral Li [9,10] and, hence, these lines are due to single-K-shell excitation. The observed double-K-shell vacancy lines, however, are nearly all associated with configurations in Li^+. This latter fact is significant because it means that K-shell ionization plus K-shell excitation events are much more likely than those due to double-K-shell excitation. The only evidence for double-K-shell-excitation is the $2s2p^2\,^2D$ configuration centered near 78 eV.

The most significant features of the double-K-shell-vacancy part of the spectra are (1) the large intensities observed for the $2s^2\,^1S$ and $2s3s\,^3S$ configurations, and (2) the fact that the $2s3s\,^3S$ intensity is larger than the

$2s^2\,{}^1S$ intensity. As will be discussed below, these large ${}^1S$ and ${}^3S$ intensities are due principally to *e-e* transitions. Particularly significant is the large intensity observed for the $2s3s\,{}^3S$ configuration, which is attributed to a three-electron transition.

To explain the large intensities observed for the $2s^2\,{}^1S$ and $2s3s\,{}^3S$ states, we consider again Fig. 1. The left side shows ionization, due to an *n-e* interaction, occurring via the dipole transition $1s \rightarrow \varepsilon p$. In principle, the monopole transition $1s \rightarrow \varepsilon s$ can also occur, but its probability is expected to be less than 2% of that for the dipole transition [20,23]. The right side of Fig. 1 shows the subsequent excitation possibilities that are expected to dominate. The most probable excitation via the *n-e* interaction is the dipole transition $1s \rightarrow 2p$ [23], giving rise to the $2s2p\,{}^3P$ state (see Fig. 2). The monopole transition $1s \rightarrow 2s$ could also occur due to the *n-e* interaction, but its probability is negligible as can be seen from Fig. 2 by comparing the intensities of the single-*K*-shell-vacancy $1s2s^2\,{}^2S$ and $1s2s2p\,{}^2P$ transitions. The

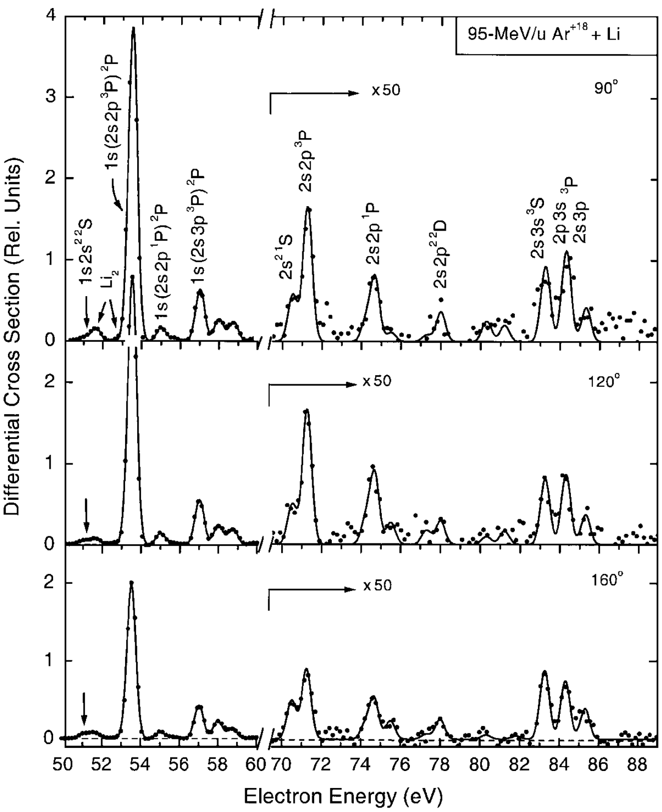

FIG. 2. High-resolution doubly differential Auger-electron-emission spectra, for emission angles of 90°, 120°, and 160°, for 95 MeV/nucleon Ar^{18+} ions colliding with atomic Li. Single-*K*-shell excitation transitions are in the range 50–60 eV, and double-*K*-shell-vacancy transitions are in the range 70–85 eV (to reference these latter energies to the ground state of neutral Li, the value of 81.03 eV must be added to the observed electron emission energies). The most prominent single- and double-*K*-shell excited-state configurations are indicated. The large intensities observed for $2s^2\,{}^1S$ and $2s3s\,{}^3S$ are particularly significant as discussed in the text.

probability for a $1s \rightarrow 3s$ transition via the *n-e* interaction would be smaller still. Thus, the large $2s^2\,{}^1S$ and $2s3s\,{}^3S$ double-*K*-shell-vacancy intensities can come about only from the *e-e* interaction.

It is mentioned here that the $2s2p\,{}^3P$ state could also have a contribution from the *e-e* interaction. This could take place if the ionized εp electron interacts with the remaining $1s$ electron, exciting it to $2p$ while simultaneously giving up its $l = 1$ angular momentum to become a continuum electron with $l = 0$, i.e., $(1s \rightarrow \varepsilon p) \Rightarrow (\varepsilon p + 1s \rightarrow 2p + \varepsilon\text{'s})$, thereby producing the $2s2p\,{}^3P$ configuration. If this dielectronic contribution to $2s2p\,{}^3P$ could be separated from the *n-e* contribution, then it would be possible to experimentally distinguish dielectronic processes from shake, since the $2s2p\,{}^3P$ state cannot be produced by shake.

Estimates for the formation of the $2s^2\,{}^1S$ and $2s3s\,{}^3S$ configurations can be obtained from the product of the Li $1s$ single ionization cross section and the corresponding shake probabilities. The shake probabilities can be calculated within the sudden approximation from the overlap of the initial-state neutral Li and the final-state Li^+ wave functions. Using the Grant atomic structure code [24], we obtain the overlap integral probabilities listed in Table I. By apportioning the cross section for the single ionization of a Li $1s$ electron (5.7×10^{-17} cm^2, from Table I) statistically between the $1s2s\,{}^1S$ and the $1s2s\,{}^3S$ states, the calculated value of 0.0067 for $|\langle 1s_{\rm Li}\,|\,2s_{\rm Li^+}\rangle|^2$, along with the subsequent branching into $2s$ or $3s$ in Li^+ (0.55 or 0.41, respectively), gives predicted intensities for $2s^2\,{}^1S$ and $2s3s\,{}^3S$ which are generally consistent with the observed intensities of these lines (not shown). These

TABLE I. Upper part: Total cross sections, obtained by integrating over angle, for single-*K*-shell excitation, single-*K*-shell ionization (from Ref. [20]), and double-*K*-shell vacancy production for the spectra shown in Fig. 2. To perform the integration, a $\sin^2\theta$ dependence was assumed for the *P* states and an isotopic dependence for the *S* states. The absolute uncertainties for the single-*K*-shell cross sections are estimated to be about ±15%, and for the double-*K*-shell cross section about ±25%. Lower part: Relevant overlap integrals for the formation of the double-*K*-shell vacancy *S* states observed in Fig. 2 (see text).

	Expt.	Theo.
Cross sections		
Single *K* excitation	133×10^{-19} cm^2	130×10^{-19} cm^2 [a]
Single *K* ionization	570×10^{-19} cm^2 [b]	⋯
Double *K* vacancy	2.9×10^{-19} cm^2	⋯
Overlap integrals		
$\lvert\langle 1s_{\rm Li}\,\vert\,2s_{\rm Li^+}\rangle\rvert^2$	⋯	0.0067 [c]
$\lvert\langle 1s_{\rm Li}\,\vert\,3s_{\rm Li^+}\rangle\rvert^2$	⋯	0.000 48 [c]
$\lvert\langle 2s_{\rm Li}\,\vert\,2s_{\rm Li^+}\rangle\rvert^2$	⋯	0.55 [c]
$\lvert\langle 2s_{\rm Li}\,\vert\,3s_{\rm Li^+}\rangle\rvert^2$	⋯	0.41 [c]

[a] From Ref. [23].
[b] From Ref. [20].
[c] From Ref. [24].

comparisons between experiment and theory will be discussed in more detail in a later paper.

It is important to note that the ionization-excitation process leading to the $2s3s\,^3S$ state is a three-step process involving one n-e interaction (ionization) followed by two e-e interactions (excitation). To our knowledge, direct evidence for such a process has not been previously observed in ion-atom collisions.

To understand how the correlated three-electron transition may occur, we consider again Fig. 1. Since the $2s3s\,^3S$ configuration is a triplet, it can only result from the intermediate $1s2s\,^3S$ configuration because this internal rearrangement cannot change the spin angular momentum of the system. For the same reason, this latter state cannot produce the $2s^2$ configuration due to "Pauli blocking." Thus, the $2s3s\,^3S$ configuration can come about only by means of a direct $1s \rightarrow 3s$ e-e transition (not shown in the figure), or by an exchange shake mechanism where the $1s$ electron goes to $2s$ and *simultaneously* the $2s$ electron is "pushed" to the $3s$ level. Based on the overlap integral of the $1s$ and $3s$ wave functions, i.e., $|\langle 1s_{\mathrm{Li}} \mid 3s_{\mathrm{Li}^+}\rangle|^2$, the probability for the direct mechanism is very small (<0.0005), and so it is likely that the exchange mechanism dominates.

In summary, by resolving individual excited-state configurations in the production of hollow Li, it was possible to identify spectral features associated specifically with the e-e interaction. The total e-e contribution to double-K-shell-vacancy production from these features, i.e., the sum of the $2s^2\,^1S$ and $2s3s\,^3S$ configurations, is comparable to the remaining double-K-shell-vacancy intensity. Furthermore, the presence of a strong $2s3s\,^3S$ configuration provides direct evidence for a three-electron transition involving correlation. Our calculations indicate that the ^1S and ^3S states are produced mainly by shake, rather than by dielectronic, processes. On the other hand, dielectronic processes could contribute to the formation of the $2s2p\,^3P$ state, and further studies may permit an unambiguous identification of this manifestation of the e-e interaction.

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[1] See, for example, J. H. McGuire, *Introduction to Dynamic Correlation* (Cambridge University Press, Cambridge, England, 1997).
[2] B. Crasemann, Comments At. Mol. Phys. **22**, 163 (1989).
[3] J. H. McGuire, Phys. Rev. Lett. **49**, 1153 (1982); Phys. Rev. A **36**, 1114 (1987).
[4] N. Stolterfoht, Nucl. Instrum. Methods Phys. Res., Sect. B **53**, 477 (1991).
[5] F. Martín and A. Salin, Phys. Rev. A **54**, 3990 (1996).
[6] J. F. Reading and A. L. Ford, in *Electronic and Atomic Collisions, Invited Papers*, edited by H. B. Gilbody, W. R. Newell, F. H. Read, and A. C. H. Smith (North-Holland, Amsterdam, 1988), pp. 693–698.
[7] B. Crasemann, J. Phys. (Paris) **48**, C9-389 (1987).
[8] See, for example, J. H. McGuire, N. Berrah, R. J. Bartlett, J. A. R. Samson, J. A. Tanis, C. L. Cocke, and A. S. Schlachter, J. Phys. B **28**, 913 (1995).
[9] P. Ziem, R. Bruch, and N. Stolterfoht, J. Phys. B **8**, L480 (1976).
[10] M. Rødbro, R. Bruch, and P. Bisgaard, J. Phys. B **12**, 2413 (1979).
[11] L. Journel *et al.*, Phys. Rev. Lett. **76**, 30 (1996).
[12] D. Cubaynes *et al.*, Phys. Rev. Lett. **77**, 2194 (1996).
[13] S. Diehl *et al.*, J. Phys. B **30**, L595 (1997).
[14] Y. Azuma *et al.*, Phys. Rev. Lett. **79**, 2419 (1997).
[15] J. A. Tanis, J.-Y. Chesnel, F. Frémont, M. Grether, B. Skogvall, B. Sulik, M. Tschersich, and N. Stolterfoht, Phys. Rev. A **57**, R3154 (1998).
[16] A. Müller *et al.*, Phys. Rev. Lett. **63**, 758 (1989).
[17] H. A. Bethe, Ann. Physics (Leipzig) **5**, 325 (1930).
[18] N. Stolterfoht, Phys. Rev. A **48**, 2980 (1993).
[19] N. Stolterfoht *et al.*, Phys. Rev. A **48**, 2986 (1993).
[20] N. Stolterfoht *et al.*, Phys. Rev. Lett. **80**, 4649 (1998); N. Stolterfoht *et al.*, Phys. Rev. A **59**, 1262 (1999).
[21] N. Stolterfoht, Z. Phys. **248**, 81 (1971); N. Stolterfoht *et al.*, Europhys. Lett. **4**, 899 (1987).
[22] N. Stolterfoht, X. Husson, D. Lecler, R. Köhrbrück, B. Skogvall, S. Andriamonje, and J. P. Grandin, in *Proceedings of the Seventeenth International Conference on the Physics of Electronic and Atomic Collisions, Brisbane, Australia, 1991, Abstracts of Contributed Papers*, edited by W. R. MacGillivray, I. E. McCarthy, and M. C. Standage (AIP, New York, 1991), p. 393.
[23] Calculations were performed using a Born approximation code provided to us by A. Salin (unpublished).
[24] K. G. Dyall, I. P. Grant, C. T. Johnson, F. A. Parpia, and E. P. Plummer, Comput. Phys. Commun. **55**, 425 (1989).