

LETTER TO THE EDITOR

Inner-shell excitation of lithium by fast charged projectiles

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Abstract. Cross sections for inner-shell and outer-shell excitations of lithium atoms in collisions with fast charged projectiles have been calculated. For these excitation processes, the relative importance of the first- and second-order mechanisms, the dependence of the cross sections on the sign of the projectile charge, as well as the effect of electron correlations in the initial and final state have been investigated. The calculated results are compared with experimental data. Correlations are found to be essential in explaining the large ratio between cross sections for an excitation to the $1s(2s2p^3P)^2P_a$ and the $1s(2s2p^1P)^2P_b$ states.

The role of electron correlations in one- and two-electron transitions has mainly been investigated for helium during the last decade [1–5]. In the case of this target, it was found that the cross sections for double ionization and ionization–excitation with antiprotons or electrons, respectively, are larger by a factor of about two, over a wide range of velocities, compared with the cross sections from equivelocity proton impact [6, 7]. This enhancement of the cross sections, dependent upon the charge of the projectile, has been discussed in several papers [8–12]. It seems to arise from the interference between the first-order and second-order processes and was found to be strongly sensitive to electron correlations. For double excitations of helium, in contrast, there is no such clear experimental evidence of the dependence of the cross sections on the sign of the projectile charge, since older experiments [13] have rather large errors and refined data [14] are only available for protons. Thus, while the Born approximation with uncorrelated wavefunctions yields generally good results for one-electron transitions, second-order processes and electron correlations are important to describe two-electron transitions.

In the present letter, we investigate similar excitation processes for the more complex lithium atom. Although we consider both outer- and inner-shell excitations, we will mainly focus on the creation of inner-shell holes which lead to autoionizing states. Such autoionizing states of lithium have been identified for a long time in various Auger spectra [15, 16]. The absolute cross sections for these autoionizing states, however, have been measured only recently by Tanis *et al* [17–19] for single and double K-shell vacancy production. Below, we compare our results with these experimental cross sections and, in particular, investigate the effect of electron correlations on these transitions. We also study the dependence of the cross sections on the sign of the projectile charge.

Our calculations were carried out within the semiclassical approximation of a well defined impact parameter. In this approximation, the projectile moves along a classical, straight-line trajectory. Then, the time-dependent Schrödinger equation needs to be considered only for the

electron system. We apply perturbation theory through second order for which the first- and second-order transition amplitudes are given by

$$a^{(1)} = -i \int_{-\infty}^{+\infty} dt e^{i(E_f - E_i)t} \langle f | V(t) | i \rangle \quad (1)$$

$$a^{(2)} = - \sum_k \int_{-\infty}^{+\infty} dt e^{i(E_f - E_k)t} \langle f | V(t) | k \rangle \int_{-\infty}^t dt' e^{i(E_k - E_i)t'} \langle k | V(t') | i \rangle. \quad (2)$$

Here i represents the initial state, k the intermediate and f the final state of the electronic system, E_i , E_k and E_f are the corresponding energies, and $V(t)$ represents the time-dependent projectile–electron interaction. In order to describe the initial and final states, we use correlated (multiconfiguration) wavefunctions as recently described for a two-electron system in [5]. Though the generalization of these formulae is also straightforward for three-electron systems like the lithium atom, the expressions are more complex in detail, since the spin-dependent part of the wavefunctions cannot be separated for more than two electrons. In this case we start directly from a representation of the atomic states in terms of Slater determinants

$$\begin{aligned} i &= \sum_i c_i \det |\phi_{a_i}(r_1) \phi_{b_i}(r_2) \phi_{c_i}(r_3)| \\ &= \frac{1}{\sqrt{3!}} \sum_i c_i \sum_P (-1)^P P \phi_{a_i}(r_1) \phi_{b_i}(r_2) \phi_{c_i}(r_3) \end{aligned} \quad (3)$$

$$\begin{aligned} f &= \sum_j d_j \det |\phi'_{a_j}(r_1) \phi'_{b_j}(r_2) \phi'_{c_j}(r_3)| \\ &= \frac{1}{\sqrt{3!}} \sum_j d_j \sum_{P'} (-1)^{P'} P' \phi'_{a_j}(r_1) \phi'_{b_j}(r_2) \phi'_{c_j}(r_3) \end{aligned} \quad (4)$$

and use the techniques known for many-electron atoms. In equations (3) and (4), the one-electron functions ϕ and ϕ' denote different sets of spin-orbitals in the initial and final states, and P and P' respectively denote the permutation of these orbitals. In the present computations, different basis sets for the initial and final states have been applied in order to improve the representation of these states and, at the same time, to keep the expansion feasible. Variational calculations have been applied to obtain the expansion coefficients c_i and d_j of the Slater determinants. As the second-order amplitude, however, includes a summation over a complete set of intermediate states, we restricted this summation to those determinants which just differ from the initial and final configuration by a single replacement of electrons. Although this computational scheme neglects correlations in the intermediate states it seems to be justified for fast collisions [4].

By taking into account the fact that the perturbation is a sum of three individual projectile–electron interactions

$$V(t) = \sum_{l=1}^3 V_l(t), \quad (5)$$

the first-order amplitude (1) can be written in terms of overlap integrals and one-electron amplitudes as

$$\begin{aligned} a^{(1)} &= -\frac{i}{3!} \sum_{l=1}^3 \sum_{i,j} \sum_{P,P'} c_i d_j^* (-1)^{P+P'} P' P \langle \phi'_{a_j} | \phi_{a_i} \rangle \langle \phi'_{b_j} | \phi_{b_i} \rangle \\ &\quad \times \int_{-\infty}^{+\infty} dt e^{i(E_f - E_i)t} \langle \phi'_{c_j} | V_l(t) | \phi_{c_i} \rangle. \end{aligned} \quad (6)$$

Similarly, the second-order amplitude is given by

$$a^{(2)} = -\frac{i}{3!} \sum_{l,l' \neq l} \sum_{i,j} \sum_{P,P'} c_i d_j^* (-1)^{P+P'} P' P \langle \phi'_{a_j} | \phi_{a_i} \rangle \langle \phi'_{c_j} | \phi_{c_i} \rangle \langle \phi'_{b_i} | \phi_{b_j} \rangle \\ \times \int_{-\infty}^{+\infty} dt e^{i(E_f - E_k)t} \langle \phi'_{b_j} | V_l(t) | \phi'_{b_i} \rangle \int_{-\infty}^t dt' e^{i(E_k - E_i)t'} \langle \phi_{c_j} | V_{l'}(t') | \phi_{c_i} \rangle. \quad (7)$$

Here, we neglected the terms containing two interactions with the same electron, because the second-order process of a single electron is insignificant for fast collisions.

The excitation cross section is obtained as an integral of the square of the amplitude over the impact parameter

$$\sigma = 2\pi \int B dB |a^{(1)} + a^{(2)}|^2. \quad (8)$$

Since the first-order amplitude $a^{(1)}$ is proportional to Z_p the charge of the projectile, and $a^{(2)}$ is proportional to Z_p^2 , an interference term which is proportional to Z_p^3 may arise in the total cross section [8]. This term causes the dependence of the cross sections on the sign of the projectile charge, however, it gives rise to a significant contribution only if the time-ordered part of the second-order amplitude [20] has the same order of magnitude as the first-order amplitude. Therefore, if this is not the case, the cross section does not depend significantly on the sign of the projectile charge.

To obtain insight into the effect of electron correlations on the cross sections (taken into account by the use of multiconfiguration wavefunctions in the initial and the final states), we carried out calculations in three different approximations: (1) using single-configuration (Hartree–Fock) wavefunctions (SC); (2) using multiconfiguration wavefunctions with a limited orbital basis up to the principal quantum number $n = 2$ (MC2); and (3) using multiconfiguration wavefunctions with an orbital basis up to $n = 3$ (MC3). For the given excitations of lithium atoms, this already results in the approximation MC2 in a wavefunction expansion of 38 determinants for the ground state and of 52 and 46 determinants for the odd- and even-parity excited states, respectively. In the approximation MC3, the corresponding expansions then include 646, 798 and 950 determinants. Due to the occurrence of open shells, most of the states must be described by a linear combination of several determinants even in the single-configuration approximation, a serious difficulty which rapidly increases if more open shells get involved.

We first applied our method to study outer-shell excitations of lithium by fast proton and antiproton impact (or by electrons with equivalent velocities). Figure 1 represents our calculated cross sections for the excitation of the lowest-lying $1s^2 2p^2 P$ state using the three different approximations along with the recommended values of Wutte *et al* [21] and some theoretical [22] and experimental [23] data for proton projectiles. In this case, the first-order dipole excitation process clearly dominates the second-order mechanism and, thus, the total cross sections as obtained for positive and negative projectiles are practically the same. For these outer-shell excitations, the use of multi-configuration wavefunctions considering orbitals up to $n = 2$ has insignificant effect and the inclusion of configurations up to $n = 3$ has increased the cross sections only slightly.

This situation changes significantly if we consider inner-shell excitations. In these cases the use of correlated wavefunctions for representing the initial and final states may dramatically change the values of the cross sections for some of the excited states.

Experimentally, the most recent and complete data for the excitation of the autoionizing states of lithium atoms [18, 19] are available for 95 MeV u^{-1} Ar^{18+} projectiles. For this reason, below we present our results for the inner-shell excitation particularly for collisions with this projectile.

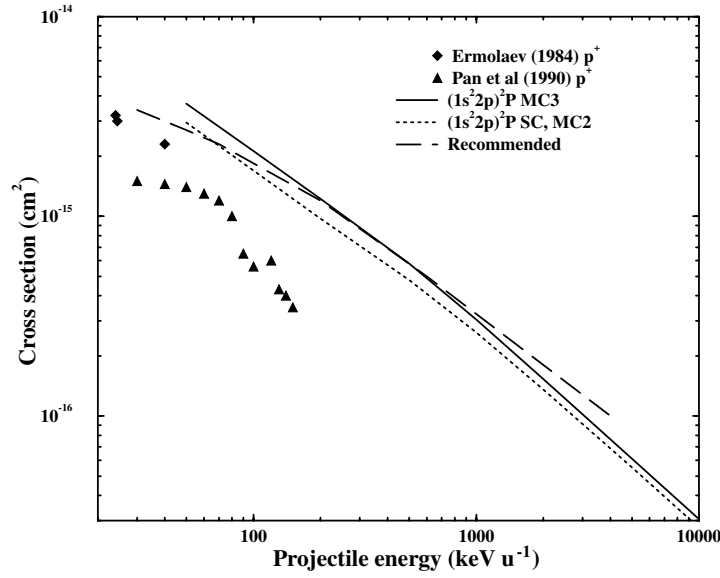


Figure 1. Cross sections for the excitation of lithium atoms to the $1s^22p$ state by proton impact as a function of the projectile energy. Our results are calculated in single-configuration (SC) and multiconfiguration (MC2 and MC3) approximations and are compared with other theoretical [22] and experimental [23] data, as well as with the recommended cross sections of Wutte *et al* [21].

The most remarkable effect of correlation arises for an excitation of the $1s(2s2p^3P)^2P_a$ and $1s(2s2p^1P)^2P_b$ states. If no correlation is taken into account beyond a single-configuration approach, these states are linear combinations of the Slater determinants

$$\begin{aligned}
 1s(2s2p^3P)^2P_a \left(+\frac{1}{2}\right) &= \frac{1}{\sqrt{6}} \det \left| 1s' \left(+\frac{1}{2}\right) 2s' \left(+\frac{1}{2}\right) 2p' \left(-\frac{1}{2}\right) \right| \\
 &+ \frac{1}{\sqrt{6}} \det \left| 1s' \left(+\frac{1}{2}\right) 2s' \left(-\frac{1}{2}\right) 2p' \left(+\frac{1}{2}\right) \right| \\
 &- \sqrt{\frac{3}{2}} \det \left| 1s' \left(-\frac{1}{2}\right) 2s' \left(+\frac{1}{2}\right) 2p' \left(+\frac{1}{2}\right) \right|
 \end{aligned} \quad (9)$$

$$\begin{aligned}
 1s(2s2p^1P)^2P_b \left(+\frac{1}{2}\right) &= \frac{1}{\sqrt{2}} \det \left| 1s' \left(+\frac{1}{2}\right) 2s' \left(+\frac{1}{2}\right) 2p' \left(-\frac{1}{2}\right) \right| \\
 &- \frac{1}{\sqrt{2}} \det \left| 1s' \left(+\frac{1}{2}\right) 2s' \left(-\frac{1}{2}\right) 2p' \left(+\frac{1}{2}\right) \right|,
 \end{aligned} \quad (10)$$

where the numbers in parentheses denote the spin states of the electrons. The coefficients for the determinants have been deduced by coupling successively the spins of the three electrons. In these formulae we neglect the orbital momentum of the p electron and spin-orbit coupling, but that would not change our conclusion. In contrast, the ground state in this approximation is simply obtained from

$$1s^22s^2S \left(+\frac{1}{2}\right) = \det \left| 1s \left(+\frac{1}{2}\right) 1s \left(-\frac{1}{2}\right) 2s \left(+\frac{1}{2}\right) \right|. \quad (11)$$

When applying formula (6) for this special case of uncorrelated transition amplitudes to the excited states above, we just take into account the direct $1s \rightarrow 2p$ (dipole) transition and neglect the indirect $2s \rightarrow 2p$ transition which is accompanied by a $1s \rightarrow 2s$ shake-up process. Moreover, suppose that a spin-flip is forbidden for the individual electrons, only those determinants with

Table 1. Mixing coefficients of the Slater determinants for the $1s(2s2p^3P)^2P$ and $1s(2s2p^1P)^2P$ states, denoted by 2P_a and 2P_b respectively, as obtained from correlated (multiconfiguration) and uncorrelated computations. To classify the individual determinants in the expansion we display the signs of the (three) spins and the value of m_l for the p electron, e.g. $-++(0)$ denotes $\det |1s(-\frac{1}{2}) 1s(+\frac{1}{2}) 2p(0, +\frac{1}{2})|$.

State (J, M_J)	Coefficients of the determinants correlated/uncorrelated					
	$-++(0)$	$+-+(+1)$	$+-+(0)$	$+-(-+1)$	$++-(0)$	$--+(+1)$
$^2P_a(\frac{3}{2}, \frac{1}{2})$	-0.641/-0.667	-0.130/-0.236	0.178/0.333	0.448/0.471	0.470/0.333	-0.337/-0.236
$^2P_b(\frac{3}{2}, \frac{1}{2})$	-0.167/0	-0.451/-0.408	0.640/0.577	0.120/0	0.475/0.577	0.338/0.408
$^2P_a(\frac{1}{2}, \frac{1}{2})$	0.460/0.471	-0.187/-0.333	-0.127/-0.236	0.643/0.667	-0.322/-0.236	-0.463/-0.333
$^2P_b(\frac{1}{2}, \frac{1}{2})$	0.109/0	-0.638/-0.577	-0.453/-0.408	0.158/0	0.338/0.408	0.482/0.577
	$++-(+1)$	$-++(+1)$	$+-+(+1)$			
$^2P_a(\frac{3}{2}, \frac{3}{2})$	0.567/0.408	-0.793/-0.816	0.209/0.408			
$^2P_b(\frac{3}{2}, \frac{3}{2})$	-0.579/-0.707	-0.202/0	0.787/0.707			

the same spin-state of the 2s electron as in the initial state remain in the expansion of the final states. Note that the one-electron amplitude then simply becomes

$$a_{1a,b}(1s \rightarrow 2p) = \int_{-\infty}^{+\infty} dt e^{i(E_{a,b}-E_i)t} \langle 2p' | V_1(t) | 1s \rangle, \quad (12)$$

while the corresponding amplitudes of the two excited states are

$$a^{(1)}[1s(2s2p^3P)^2P_a] = \sqrt{\frac{3}{2}} \langle 1s' | 1s \rangle \langle 2s' | 2s \rangle a_{1a}(1s \rightarrow 2p) \quad (13)$$

$$a^{(1)}[1s(2s2p^1P)^2P_b] = \sqrt{\frac{1}{2}} \langle 1s' | 1s \rangle \langle 2s' | 2s \rangle a_{1b}(1s \rightarrow 2p), \quad (14)$$

with E_a and E_b being the total energies of these excited states. Following this (uncorrelated) single-configuration approximation, the ratio of the cross sections for excitation into the triplet and the singlet states should always be around 3, because the small energy difference between the two excited states leads to almost the same one-electron amplitude (they differ by 0.2%). Experimentally, however, a ratio of 21 was found, much larger than that derived from such a simple approximation [19].

The configuration interaction among these inner-shell excited levels of lithium clearly changes the representation of the atomic states, i.e. the coefficients in the determinant expansion relative to the single-configuration approximation. An improved, correlated computation, which includes active sets of orbitals up to $n = 2$, leads to the coefficients displayed in table 1; they have to be compared with the (uncorrelated) coefficients as obtained from the pure coupling of the spins of the electrons (cf equations (9) and (10)) along with the orbital momentum of the 2p electron. These changes in the coefficients also lead to quite a remarkable modification of the ratio of the cross sections for excitations into the 2P_a and 2P_b states which increases to more than 15. This ratio has to be compared with the experimental value of 21 as listed in table 2. We also observed that the inclusion of the $n = 3$ orbitals into the active set further lowers the total cross section for the two states close to its experimental value. These correlations, however, still increase the ratio of the individual cross sections significantly to almost double those of the experimental ones. Our results therefore suggest that even larger wavefunction expansions are needed to obtain a satisfactory agreement with experiment. But

Table 2. Cross sections (in 10^{-20} cm^2) for the excitation of the $1s(2s2p^3P)^2P$ (2P_a) and $1s(2s2p^1P)^2P$ (2P_b) states of lithium by $95 \text{ MeV u}^{-1} \text{ Ar}^{18+}$ projectiles, calculated in three different approximations and compared with experiment. SC denotes a single configuration approximation neglecting the shake mechanism, while SCSH includes the shake mechanism. MC2 and MC3 denote multiconfiguration calculations at two different levels of complexity.

	SC	SCSh	MC2 ($n \leq 2$)	MC3 ($n \leq 3$)	Exp. [19]
2P_a	981	838	1310	1222	950
2P_b	326	278	85.5	27.5	45
$^2P_a + ^2P_b$	1307	1116	1395	1249	995
$^2P_a/{}^2P_b$	3.01	3.01	15.3	44.1	21

Table 3. Calculated cross sections (in 10^{-20} cm^2) for the excitation of lithium by $95 \text{ MeV u}^{-1} \text{ Ar}^{18+}$ projectiles, using configuration interaction (MC3) wavefunctions in comparison with the experimental data and results from a Born approximation [19].

State	First order	second order	Present theory	Exp. [19]	Theory [19]
$(1s2s^2)^2S$	22.2	1.35	23.6	12.7	21
$1s(2s2p^3P)^2P_a$	1222	—	1222	950	—
$1s(2s2p^1P)^2P_b$	27.5	—	27.5	45	—
$^2P_a + ^2P_b$	1249	—	1249	995	972
$(1s2p^2)^2D$	0.20	11	12.5	9.49	18.3
$(1s2s^3S)3s^2S$	5.95	3.32	9.22	—	—
$1s2s3p^2P$	138 ^a	—	138 ^a	173	166
$(1s2s^3S)3d^2D$	2.02	2.45	4.54	—	—
$(1s2s^1S)3s^2S$	1.65	0.07	1.74	—	—
$1s2s4p^2P$	42 ^a	—	42 ^a	66	82
$(1s2p^2)^2S$	20.9	0.87	21.9	28.1	4.8
$1s2s5p^2P$	20.9 ^a	—	20.9 ^a	—	—

^a Calculated in a single-configuration approximation.

the presently applied expansions already show, however, that the sum of the cross sections appears less sensitive to the electron correlations than their ratio does.

Table 3 presents our results for the inner-shell excitation of lithium by $95 \text{ MeV u}^{-1} \text{ Ar}^{18+}$ projectiles. In order to specify the importance of the first- and second-order mechanisms, their individual contributions are listed separately. Excluding the np excited states with $n \geq 3$, our final results are obtained within the approximation MC3. Our theoretical cross sections show good agreement with the experimental data. Relative to the theoretical data from the Born approximation by Tanis *et al* [19] (i.e. by applying single-configuration wavefunctions), the largest difference arises for the excitation of the $(1s2p^2)^2S$ state, most likely owing to the strong configuration interaction of this configuration with the $(1s2s^2)^2S$ one. Excepting this, and the already discussed case of the $1s2s2p^2P$ states, the inclusion of electron correlations in the calculations does not cause very important modifications to the cross sections.

In order to investigate the dependence of the inner-shell excitation cross sections on the energy and on the charge sign of the projectile, we have also performed the calculations for proton and antiproton projectiles, although there are no available experimental data to compare with.

Figure 2 displays results for the excitation to the $1s2s2p^2P$ states. Since the first-order process dominates the overall excitation into these states, the cross sections do not depend much on the sign of the projectile charge. The ratio of the cross sections to the two states becomes less influenced by electron correlations as the projectile energies increase.

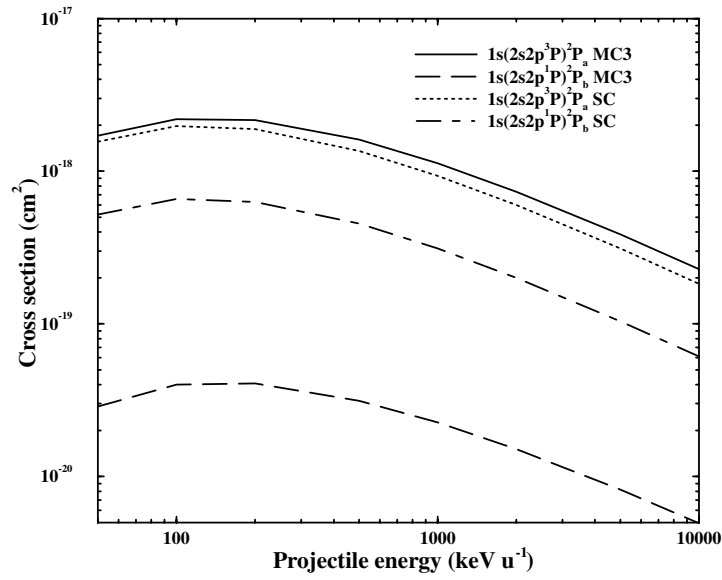


Figure 2. Cross sections for the excitation of the $1s(2s2p^3P)^2P_a$ and $1s(2s2p^1P)^2P_b$ states of lithium by proton impact, as a function of the projectile energy, calculated in single configuration (SC) and configuration interaction (MC3) approximations.

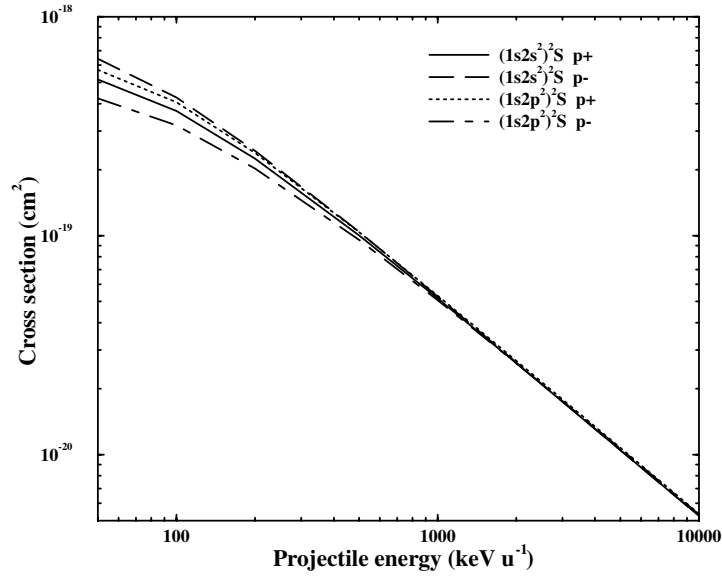


Figure 3. Cross sections for the excitation of the $(1s2s^2)^2S$ and $(1s2p^2)^2S$ states of lithium by proton and antiproton impact, as a function of the projectile energy.

Figures 3–5 display the energy dependence of the cross sections for the excitation of the $(1s2s^2)^2S$, $(1s2p^2)^2S$, $(1s2s^3S)3s^2S$, $(1s2s^1S)3s^2S$, $(1s2p^2)^2D$ and $(1s2s^3S)3d^2D$ states. Note that the interference between the first and second-order amplitudes may lead to higher cross sections for antiprotons in some cases (cf $(1s2s^2)^2S$, $(1s2s^3S)3s^2S$, $(1s2p^2)^2D$), while

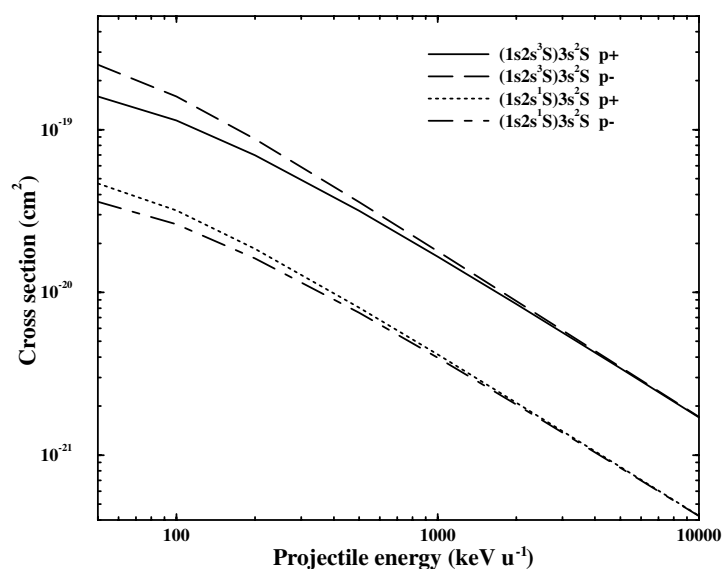


Figure 4. As figure 3, but for excitation of the $(1s2s^3S)3s^2S$ and $(1s2s^1S)3s^2S$ states.

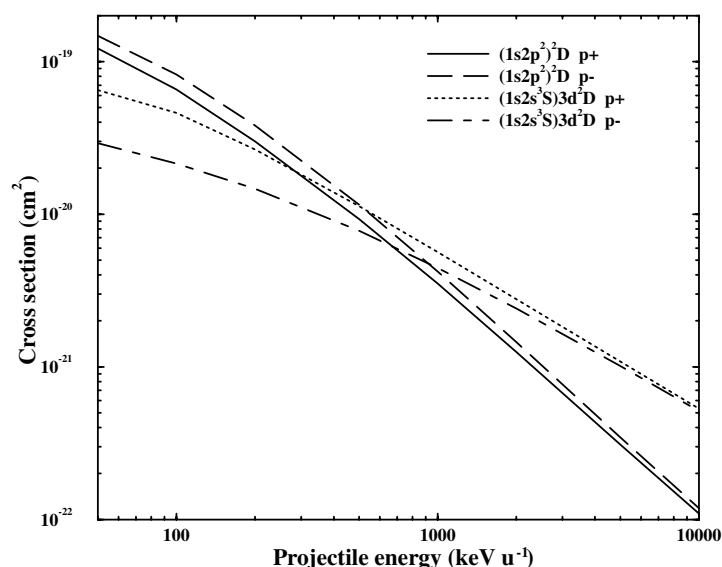


Figure 5. As figure 3, but for excitation of the $(1s2p^2)^2D$ and $(1s2s^3S)3d^2D$ states.

they are lower than for protons in other cases (cf $(1s2p^2)^2S$, $(1s2s^1S)3s^2S$, $(1s2s^3S)3d^2D$). Experimental investigations on the double ionization and the ionization–excitation for helium [6, 7] and for the hydrogen molecule [24] showed that cross sections for negative projectiles are typically higher than those for positive projectiles with equal velocity over a wide velocity range. Theoretical calculations, however, suggest that for some two-electron transitions in helium—where no experimental data are available—this should not be always the case [25, 26]. It therefore seems that for inner-shell excitations of lithium also, the sign of the interference contribution to the cross section depends on the final excited state.

It is interesting to remark upon the different behaviour of the cross sections as a function of the projectile energy for the excited states represented in figure 5. While for the $(1s2p^2)^2D$ state the second-order mechanism dominates and the cross section decreases as $1/E_p^2$, the first-order contributions are dominant for excitations to the $(1s2s^3S)3d^2D$ state for which the cross section decreases roughly proportional to $1/E_p$; here, E_p denotes the projectile energy.

We conclude that our calculated cross sections for the inner-shell excitation of lithium are in satisfactory agreement with experimental data [19]. The large ratio of the cross sections for the excitation to the $1s(2s2p^3P)^2P_a$ and $1s(2s2p^1P)^2P_b$ states is due to electron correlations which were taken into account by allowing configuration mixing in the final excited states. For the first time, we also investigated the dependence of the cross sections on the sign of the projectile charge in the case of a complex system like lithium.

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