Double-electron-capture cross sections for Li^{3+} collisions with He at intermediate-to-high velocities

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(Received 11 August 2009; published 15 October 2009)

We have measured double-electron capture cross sections for Li^{3+} projectiles and He targets in the 2.0–5.5 MeV energy range (3.4–5.6 a.u. velocity range). Double-electron-capture cross sections for this collision system were measured before, but only at relatively lower energies. There is a good agreement between the three available sets of experimental data for double capture at the lower end of the present data energy interval, i.e., where the ranges from different measurements overlap. The present measurements for other collision channels that include single and double ionization, single capture, and transfer ionization corroborate the reliability of the data normalization adopted in our work. Our experimental data for double capture are compared with the corresponding theories in order to assess the relative performance of the main two strategies, the semiclassical independent particle model, and the full quantum-mechanical four-body theories. We thoroughly discuss possible reasons for the discrepancies among various theoretical predictions from these two frameworks and point out several potential paths for their improvement.

DOI: 10.1103/PhysRevA.80.042707 PACS number(s): 34.70.+e, 34.50.Fa

I. INTRODUCTION

The motivations for studying atomic collision phenomena in few-electron collision systems are twofold. From the basic physics perspective, these are suitable testing systems that already present the emergence of many-body characteristics [1,2]. Furthermore, the relative simplicity of these collisional systems alleviates the need for cumbersome and less accurate statistical theoretical approaches [3]. From the perspective of applications in other areas of physics and other sciences as well as technologies, these few-electron collisional systems are also outstanding. The associated charge-changing cross sections constitute an important part of the input databases for modeling of energy losses of heavy particles in their passage through matter as encountered in, e.g., high-temperature thermonuclear fusion [4], hot plasmas [5,6], heavy-ion therapy [7], etc.

Among the four-body collision problems, the collision channel for double capture of bare ionic projectiles X^p, with the initial charge state p, incident on two-electron atomic or ionic targets Y^q, with the initial charge state q, as symbolized by,

$$X^p + Y^q \to X^{(p-2)} + Y^{(q+2)}, \quad (1)$$

has a prominent place. Here, both electrons are transferred from the target to the projectile. Thus, *a priori* neither electron is justified to be viewed merely as a spectator during the collision. Such a circumstance *de facto* reinforces the four-body character of the system. In practice for many cases of interest, Y is initially in the ground state. However, measured cross sections that are not state selective contain the contributions from singly- and doubly-excited final states that have to be accurately taken into account. This is also the case for double capture in the Li^{3+}-He^0 collision,

$$Li^{3+} + He^0 \to Li^+ + He^{2+}, \quad (2)$$

which is presented and discussed in detail in the next sections of this paper.

The technological interest in the collision channels described by Eq. (1) is not restricted to neutral targets. For example, cross sections for the collision channel,

$$He^{2+} + Li^+ \to He^0 + Li^{3+}, \quad (3)$$

are essential parameters for the reliable application of the pellet charge exchange (PCX) diagnostic technique [8–11]. The PCX is among the several ion-beam diagnostic techniques currently used to probe magnetically confined plasmas for controlled fusion devices.

For the collision (2), there are just two sets of experimental data that are not fully in the intermediate-to-low velocity region. These sets, measured by Nikolaev *et al.* [12] as well as by Shah and Gilbody [13] in the 1–2 MeV energy range, are not well described by any theoretical method available in the literature. Higher velocity data are clearly needed as a

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TABLE I. Measured charge-changing cross sections for the Li^{3+}-He^0 collision system: double capture (DC), single capture (SC), double ionization (DI), single ionization (SI), and transfer ionization (TI). Cross sections are in the 10^{-18} cm^2 units.

E (MeV)	DC	SC	DI	SI	TI
2.0	0.38 ± 0.02	17.0 ± 0.3	15.3 ± 0.08	302 ± 8	7.59 ± 0.20
2.5	0.111 ± 0.005	9.66 ± 0.16	15.2 ± 0.14	296 ± 5	3.83 ± 0.07
3.0	0.0378 ± 0.0054	5.22 ± 0.09	11.8 ± 0.06	268 ± 3	1.96 ± 0.04
3.5	0.0136 ± 0.0020	3.05 ± 0.06	8.98 ± 0.87	238 ± 6	0.990 ± 0.028
4.0	0.0060 ± 0.0005	2.15 ± 0.14	8.63 ± 0.79	237 ± 12	0.650 ± 0.058
4.5	0.0034 ± 0.0006	1.50 ± 0.13	6.80 ± 0.73	224 ± 14	0.445 ± 0.066
5.0	0.00115 ± 0.00028	0.90 ± 0.07	5.20 ± 0.46	195 ± 19	0.250 ± 0.030
5.5	0.00047 ± 0.00016	6.6 ± 0.6	4.75 ± 0.50	188 ± 12	0.162 ± 0.019

guidance and test for various theoretical modeling. The experimental data presented in this paper can serve as a benchmark for these theoretical predictions at the intermediate-to-high velocity range.

In the present paper we report our measured cross sections in the 2.0–5.5 MeV energy range (corresponding to the incident velocities in the 3.4–5.6 range in atomic units) not only for double capture (DC) given by Eq. (2), but also for single capture (SC), double ionization (DI), single ionization (SI), and transfer ionization (TI) (Table I),

$$Li^{3+} + He \rightarrow Li^{2+} + He^{+} \quad (SC), \tag{4}$$

$$Li^{3+} + He \rightarrow Li^{3+} + He^{2+} + 2e^{-} \quad (DI), \tag{5}$$

$$Li^{3+} + He \rightarrow Li^{3+} + He^{+} + e^{-} \quad (SI), \tag{6}$$

$$Li^{3+} + He \rightarrow Li^{2+} + He^{2+} + e^{-} \quad (TI). \tag{7}$$

Cross sections for these four latter channels have already been experimentally determined at intermediate-to-high velocities and are mainly used in this paper as a check for the normalization of our double-capture data. A reliable normalization is crucial for the discussion developed in Sec. III.

This paper is organized as follows. Section II briefly describes the experimental setup used. In Sec. II, we also discuss peculiarities of the measurement of capture cross sections for collision systems with few-electron targets and high-velocity projectiles. In Sec. III we present the measured cross sections for Li^{3+}-He^0 collisions. In Sec. III, we compare the presently measured double-capture cross sections to those from several theoretical models available in the literature and put our data in the context of the current challenges for the description of four-body problems in atomic collisions. In Sec. IV, we present a summary and draw conclusions.

II. EXPERIMENTAL METHOD

The present experiments were performed at the 1.7 MV tandem accelerator of the Federal University of Rio de Janeiro. Details on the experimental setup used were reported in our previous publications [14,15] and only a brief description of the apparatus will be given here. In short, Li^- negative ions are produced at a sputtering ion source, accelerated toward a gaseous cell located at the center of the tandem accelerator, where some ions become neutralized (Li^0) or positively charged ($Li^p, 0 < p \leq 3$). The latter positive ions are further accelerated, leaving the accelerator with energies in the MeV range. In particular, the Li^{3+} beam component is magnetically deflected to the 15° line, collimated and allowed to traverse a set of electrostatic deflection plates, placed just before the collision chamber, which eliminates unwanted charge-state components. The beam crosses a gaseous jet of helium target at the center of the collision vacuum chamber. The beam emerging from the collision chamber is charge analyzed by a second parallel plate deflector. The distinct charge-state components are detected in a second chamber, placed two meters apart from the first one. Meanwhile, some of the helium atoms lose one or both electrons at the interaction region, and the He recoil ions are measured in coincidence with the projectile final charge state, using a time-of-flight (TOF) system. The cross-section data were normalized to previous measurements of Ar ionization by protons at 1.0 MeV [16]. An independent check for the normalization procedure was also performed, using data for Ar ionization by C^{3+} ions at 2.0 MeV [17]. This check agreed within ten percent with the proton normalization.

It is also worthwhile to mention that double-capture measurements include final Li^+ in ground state and singly-excited states. Only a fraction of the doubly-excited Li^+ projectiles emerging from the collision chamber is detected. If these ions decay by an Auger-like process before the analyzing parallel-plates deflector, they will be counted as Li^{2+} thus underestimating and overestimating the double- and single-capture cross sections, respectively.

In principle, double-electron-capture cross sections could also be measured with the simpler setup of a gaseous static collision cell and the growth-rate method (e.g., [18]). However, in the case of He targets and high collision velocities, coincidence measurements result in more reliable cross-section data. The reason is related to the role of a possible small amount of impurities in the He gas fed either to the gas jet (in the coincidence measurements) or to the static gas

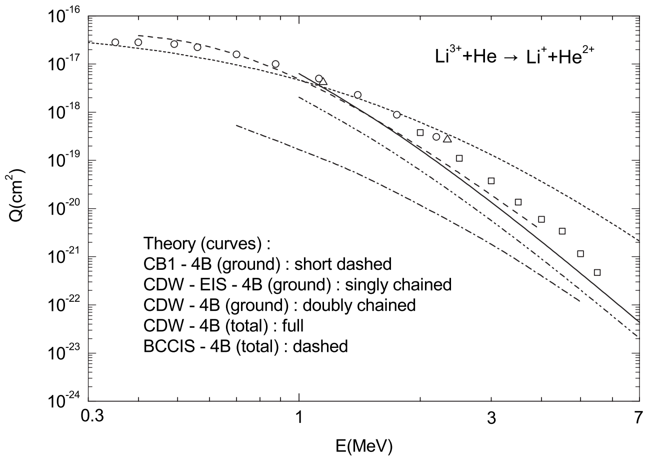

FIG. 1. Cross sections for Li^{3+} double-electron capture on He as a function of the projectile energy. Experimental data: open squares (this work); open triangles (Nikolaev *et al.* [12]); and open circles (Shah and Gilbody 1985 [13]); Theoretical models: singly-chained curve, CDW-EIS-4B (final ground state) (Gayet *et al.* [20,21]); doubly-chained curve, CDW-4B (final ground state) (Gayet *et al.* [22]); full curve, CDW-4B (final ground+excited states) (Gayet *et al.* [22]); dashed curve, BCCIS-4B (final ground+excited states) (Gosh *et al.* 2008 [23]) and dotted curve, CB1-4B (final ground state) (Belkić [24,25]).

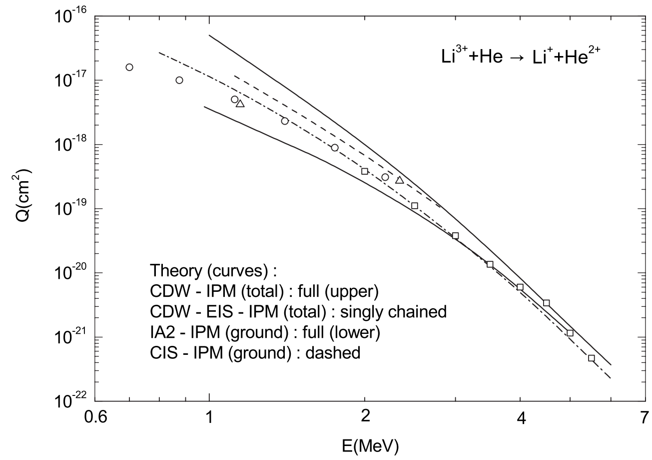

FIG. 2. Cross sections for Li^{3+} double-electron capture on He as a function of the projectile energy. Experimental data: open squares (this work); open triangles (Nikolaev *et al.* [12]); and open circles (Shah and Gilbody 1985 [13]); Theoretical models: lower full curve, IA2-IPM (final ground state) (Gravielle and Miraglia 1992 [26]); singly-chained curve, CDW-EIS-IPM (final ground+excited states) (Martinez *et al.* [27]); dashed curve, CIS-IPM (final ground state) (Ghosh *et al.* [28]) and upper full curve, CDW-IPM (final ground+excited states) (Martinez *et al.* [27]).

target (in the growth-rate method). Single-electron capture at high velocity in few-electron target such as atomic hydrogen, molecular hydrogen, and helium present small cross sections and possible target contamination are already a potential problem. For multiple-electron capture this problem is even worse. At intermediate-to-high and high velocities capture probabilities decrease fast with impact velocity. Few-electron targets He and H_2 have only electrons with the 1*s*-like orbitals and when the cross sections for one-electron capture start to decrease with velocity, as they do at high energies, the cross sections for two electrons being captured plunge, reaching very small values. On the other hand, many-electron atomic and molecular impurities at the experimental gas target have inner-shell electrons, and their capture cross sections present maxima at high velocities. Moreover, having many electrons, it is statistically easier for the projectile to probe the high-momentum part of the wave function of two target electrons in the same collision. As a result, when the growth-rate method is used, a very small amount of contaminants can result in a large overestimation of the measured cross sections. On the other hand, in coincidence TOF measurements, having different masses, these impurities do not contribute significantly to the helium signal.

III. RESULTS AND DISCUSSION

Our measured cross sections for DC by Li^{3+} incident on He are displayed in Fig. 1. Figures 2 and 3 are also concerned with DC. The present results are compared with the other experimental data available at lower impact energies and with several theoretical models to be discussed in this section. In Fig. 4 we present our data for other charge-changing collision channels, such as SI, SC, TI, and DI. Cross sections for these latter channels were previously measured and analyzed by other authors at intermediate-to-high velocities [13,19] and they will not be discussed hereafter. The present SI, SC, TI, and DI data are reported for completeness and primarily to check the reliability of our experimental setup and an independent normalization procedure.

Figure 1 for DC in Li^{3+}-He^0 collisions shows a clear consistency of our high-energy experimental data with those at intermediate energies reported earlier by Nikolaev *et al.* [12] as well as by Shah and Gilbody [13]. Regarding comparisons between experimental and theoretical findings, to avoid clut-

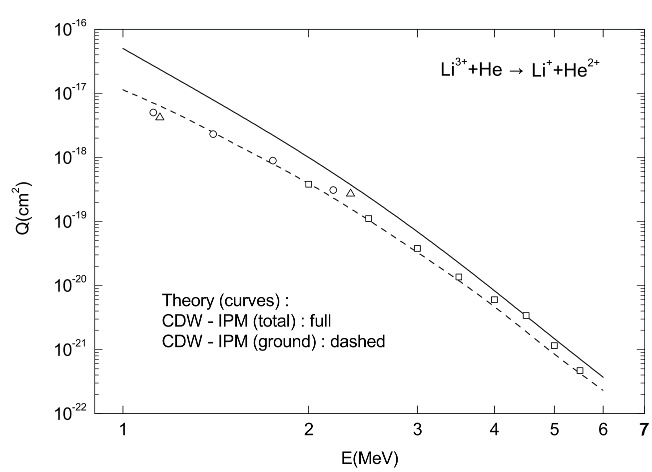

FIG. 3. Cross sections for Li^{3+} double-electron-capture on He as a function of the projectile energy. Experimental data: open squares (this work); open triangles (Nikolaev *et al.* [12]); and open circles (Shah and Gilbody 1985 [13]); Theoretical models: dashed curve, CDW-IPM (final ground state) (Martinez *et al.* [27]) and full curve CDW-IPM (final ground plus excited states) (Martinez *et al.* [27]).

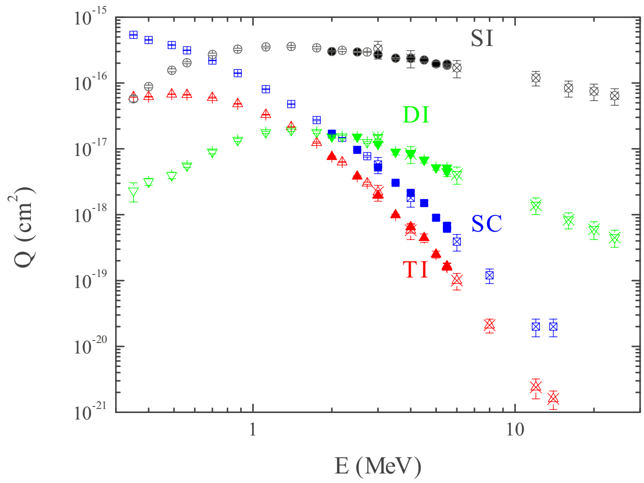

FIG. 4. (Color online) Measured total cross sections for Li^{3+}-He^0 collisions as a function of the projectile energy. Collision channels previously measured at intermediate-to-high velocities: circles (single ionization, SI); squares (single capture, SC); triangles (transfer ionization, TI); inverted triangles (double ionization, DI); References: filled symbols (this work); open symbols (Shah and Gilbody [13]); crossed symbols (Woitke *et al.* [19]).

ter, the full quantum-mechanical four-body (4B) methods and semiclassical independent particle models (IPM) are depicted at two separate figures, Figs. 1 and 2, respectively. The results from the 4B formalism shown in Fig. 1 are due to the boundary-corrected first Born (CB1-4B) [24,25], the boundary-corrected continuum intermediate states (BCCIS-4B) [23], the continuum distorted wave (CDW-4B) [22], and the continuum distorted wave initial state (CDW-EIS-4B) [21] methods. Figure 2 displays the cross sections from the corresponding IPM treatments within the continuum distorted waves (CDW-IPM) and its eikonalization (CDW-EIS-IPM) [27] and the continuum intermediate states (CIS-IPM) [28], as well as the second-order impulse approximation (IA2-IPM) [26].

All the presented 4B methods from Fig. 1 satisfy the correct boundary conditions, i.e., they possess the properly behaving wave functions at asymptotically large interparticle separations in the entrance and exit channels. Importantly, these total scattering wave functions are correctly connected to the pertinent perturbation potentials that are responsible for the transitions from the initial to the final states of the whole system under study. By contrast, the CIS method [29] and the standard impulse approximation [30,31] disobey the correct boundary conditions for any rearranging collision within both IPM and 4B formalisms. A formal relationship of CIS and IA is that the former can be deduced from the latter by means of the usual peaking approximation applied to the momentum-space bound state vector from the total scattering wave function in the impulse approximation [31]. Total cross sections available from the past, abundant literature on SC show that performances of CIS and IA are similar at impact energies at which the classical Thomas double scattering is not dominant.

A common feature seen in Figs. 1 and 2 is a large spread among the presented theories and a varying degree of agreement with the experimental data. Such discrepancies within the 4B formalism in Fig. 1 are indeed enormous and attain two orders of magnitude when comparing the CB1-4B and CDW-EIS-4B methods. Differences within one order of magnitude are also observed between the CDW-IPM and IA2-IPM as evidenced in Fig. 2 concerning the IPM treatments. When Figs. 1 and 2 are juxtaposed to each other, it follows that even at high energies, there is a significant disaccord of the cross sections obtained by the IPM and 4B methods.

At first glance, and contrary to the common expectation, it appears that certain methods in their simpler, IPM variants outperform the more elaborate, 4B formulations, by exhibiting a better agreement especially with the present high-energy experimental data in the 2–6 MeV range. This is evidently clear for the CDW-EIS-4B and CDW-EIS-IPM from Figs. 1 and 2, respectively. Contrary to this, it appears that the same conclusion does not hold true for the CDW-4B and CDW-IPM, since the former (latter) underestimate (overestimate) the present experimental data, respectively, as obvious from Figs. 1–3. Furthermore, in the 3–5 MeV energy range, Fig. 2 indicates that IA2-IPM is in good agreement with our measured cross sections. However, this is deceiving due to a twofold circumstance: (i) the matrix element due to IA1-IPM neglected in IA2-IPM is far from being negligible, as is clear from CIS-IPM in Fig. 2 (as stated, based upon the experience with SC, it is expected that the first orders of IA and CIS, i.e., IA1-IPM and CIS-IPM$\equiv$CIS1-IPM for DC also give similar cross sections at nonasymptotic impact energies), and (ii) IA2-IPM from Ref. [26] includes only the nonresonant ground-to-ground state transition $He(1s^2)\to Li^+(1s^2)$ in process (2). In short, the apparent good agreement between IA2-IPM and the present measured data at energies 3–5 MeV would not persist in a more complete computation with the inclusion of the neglected contributions from IA1-IPM and the excited states of Li^+. Note that in our experiment as well as in all the other cited measurements, the postcollisional state of Li^+ is not detected and, therefore, the theory should contain the sum of the dominantly contributing channels for the helium-like ion Li^+.

A. Critical assessment of available theoretical methods for double-electron capture

For interpretation of data from atomic scatterings, the projectile and target nuclei are permitted to be considered as structureless particles, of course. Therefore, the presently studied collision system (Li^{3+}-He^0) is a pure four-body problem. As mentioned, unlike SC, in DC neither of the two electrons of the target can be considered as being passive since they are both transferred from one center (He^{2+}) to the other (Li^{3+}) by ionization of He and formation of Li^+. Thus, our measured data should be suitable for assessing the adequacy of pure four-body theories that, in principle, are capable of describing both sequential and coherent mechanisms of DC.

In IPM for DC each electron is captured independently due to the outright neglect of the interelectron potential. These two independent encounters are modeled by the product $P_1(b)P_2(b)$ of the three-body (3B) impact parameter (b) dependent probabilities $P_1(b)$ and $P_2(b)$ for each event. The total cross section Q is obtained by integration of the composite probability $P_1(b)P_2(b)$ over all the impact parameters $(0 \le b \le \infty)$. In principle, total cross sections from IPM should tend to those from 4B methods at high impact energies. However, at intermediate and lower energies, the concept of independent capture of each electron ceases to be applicable [1,2]. This can also be inferred from Figs. 1 and 2. Nevertheless, despite the inadequacy of IPM at intermediate and lower energies, the computations within CDW-IPM [20] were needed to direct the attention toward the importance of the contributions from excited states. This was subsequently confirmed in the 4B formalism, indicating that at intermediate and smaller energies, the main contributions to DC stems from single-excited states, although the ground states of helium-like systems also provide a large contribution of about 40% [22,23,27,32]. Conversely, at high energies, DC to the ground states yields the dominant contributions.

Previous comparisons between, e.g., the CDW-4B and CDW-IPM for DC revealed a varying degree of success of each of these two treatments [1,2,20–22,27,33–41]. Thus, for H^+-He collisions, excellent agreement between measurements and CDW-4B exists [1,2,33,34] already above 80 keV, as opposed to a significant overestimation by the CDW-IPM [20]. By contrast, for He^{2+}-He collision, CDW-IPM appears to be superior to CDW-4B [1,2,20]. Regarding Li^{3+}-He^0 and B^{5+}-He collisions, large discrepancies were found between CDW-4B and CDW-IPM [20–22,27] even at relatively high energies in the MeV/amu range. Moreover, neither of the two variants of the same theory emerged with a clear advantage in comparisons with previous measurements for these two collisional systems [13,42].

Other theoretical models were also used for DC in He^{2+}-He collisions such as the four-body boundary-corrected continuum intermediate state (BCIS-4B) [1,2,43] as well as the four-body Born distorted wave (BDW-4B) approximations [1,2,35,36], yielding a close mutual agreement and comparing favorably with the experimental data for total cross sections. Purkait *et al.* [32] and Ghosh *et al.* [23] used the acronym BCCIS-4B to refer to the variant of BCIS-4B with the full Coulomb waves for the relative motions of heavy nuclei instead of the corresponding logarithmic phase asymptotes employed by Belkić [43]. These latter two treatments with the said full and asymptotic Coulomb wave functions are equivalent for heavy particle collisions and, as such, are expected to give the same total cross sections in the eikonal approximation in which the mass of the nuclei is much larger than the electronic mass (for more details, see Refs. [1,2]).

Computations in BCCIS-4B [23,32] show good agreement with the earlier experimental data on DC in He^{2+}-He and Li^{3+}-He^0 collisions. However, for the latter collision, it is seen in Fig. 1 that BCCIS-4B underestimates all our measured cross sections. Also applied to DC in the He^{2+}-He and Li^{3+}-He^0 collisional channels were the CDW-EIS-4B and CDW-EIS-IPM [21,27,41]. Here, however, while CDW-EIS-IPM is reasonably acceptable for DC due to the reliance upon the corresponding successful 3B counterpart (CDW-EIS-3B), it was found in Refs. [21,27] that CDW-EIS-4B flagrantly underestimates (by orders of magnitude) the measurements on DC in He^{2+}-He and Li^{3+}-He^0 collisions even at MeV/amu energies. This can also be seen in Fig. 1.

When compared to the experimental data, another four-body model is also known to dramatically break down at MeV/amu energies. This is the case with the CB1-4B approximation [24,25] for He^{2+}-He and Li^{3+}-He^0 collisions, although a relatively good success was achieved at intermediate impact energies according to Fig. 1 for the latter scattering (this agreement, which is based upon the ground-to-ground state transition alone, would cease to exist if the excited states of Li^+ are taken into account).

An approximate version of the IA2-IPM was tried by Gravielle and Miraglia [26] for DC in He^{2+}-He and Li^{3+}-He^0 collisions with a reasonable success relative to measurements, as seen in Fig. 2 for the latter scattering. However, such an agreement is fortuitous for the reasons (i) and (ii) given earlier in this section. The model from Ref. [26] uses the second-order propagator for a two-step collision with the inclusion of the intermediate state on-shell Green's function centered on the target and projectile nuclei. In IA2-IPM, as already emphasized, the contribution from the matrix element due to the associated first-order transition in this approximation (IA1-IPM, or simply, IA-IPM) was omitted. It was assumed [26] that IA1-IPM should yield small cross sections as in an earlier work of Crothers and McCarroll [44] who employed the independent event model (IEM) of CDW, as denoted by CDW-IEM, which is, in spirit, quite similar to CDW-IPM. However, as discussed, this assumption of Gravielle and Miraglia [26] is not supported by CIS-IPM. It is not supported by CDW-4B2 either since CDW-4B2 contains a large contribution from CDW-4B1 [41] for He^{2+}-He collisions [41]. To re-emphasize, as opposed to all the other mentioned 4B methods, IA always disregards the proper boundary conditions for DC for any scattering aggregates, similarly to the corresponding well-known unsatisfactory status of this theory for SC. Therefore, any agreement of this method with experimental data should be taken with caution because of the incorrect description of the asymptotic region of scattering where all experiments are performed.

The CDW-4B2 improves significantly CDW-4B1 for He^{2+}-He collisions and yields a good agreement with experimental data [41]. Nevertheless, this improvement may be fortuitous since the second-order propagator in the CDW-4B2 is from the ordinary distorted wave perturbation series with disconnected diagrams, as opposed to the Dodd-Greider expansion [1,2,45]. Moreover, the computations in CDW-4B2 [41] include only the on-shell part of a model two-center Green's function and retains merely the intermediate ground states centered at the projectile and target nucleus with no estimate on the contribution from the intermediate excited states and continuum. Further, CDW-4B2 [41] is concerned only with the ground-to-ground state transition $He(1s^2) \rightarrow Li^+(1s^2)$ in process (2), thus neglecting all the final excited states of Li^+ that yield a sizable contribution. As pointed out, all these drawbacks of CDW-4B2 [41] are also present in IA2-IPM [26] due to the use of the same model Green's function for intermediate states. Thus far, CDW-4B2 was not applied to Li^{3+}-He^0 collisions.

Faced with an unexpected failure of CDW-EIS-4B for DC, Martínez *et al.* [41] attempted to find an improvement in this model by following Gravielle and Miraglia [26] and thus employed the second-order transition operator via the two-center on-shell Green's function with the one-electron intermediate ground states on the projectile and target nuclei. However, the ensuing CDW-EIS-4B2 has not met with success since a huge overestimation of experimental data for DC in He^{2+}-He collisions was recorded [41]. To date, no computations were reported using CDW-EIS-4B2 for Li^{3+}-He^0 collisions.

To better comprehend the potential reasons for the varying performance of the discussed models when passing from one to another collisional system, it is necessary to highlight the key ingredient of the computations. First of all, some of the cited references on theory dealt exclusively with the ground-to-ground state DC [24,25,33–36], whereas there were studies that additionally included the contributions from a limited number of excited states [20,22,23,26,27,32]. The case of $H^+ + He(1s^2) \to H^-(1s^2) + He^{2+}$ collision is special since the negative hydrogen ion H^- has no excited states [46]. This is DC to which the first order of CDW-4B, i.e., CDW-4B1, was originally applied by Belkić and Mančev [33,34] with the conclusion cohering to an empirically established fact: this 4B model is adequate at impact energies above 80 keV.

Approximately the same lowest limit of the applicability of CDW-3B and CDW-4B is expected since this validity limit should be independent of the number of captured electrons. Other helium-like ions with nuclear charges larger than unity possess excited states and these need to be included in computations of cross sections for DC. Thus, Gayet *et al.* [22] used CDW-4B for DC with allowance for certain low-lying singly-$(1s,nl)\{n \leq 3(0 \leq l \leq n-1)\}$ and the first doubly-$(2l,2l')\{l=0,1 l'=0,1\}$ excited helium-like states in the exit channels of $He^{2+} + He \to He + He^{2+}$, $Li^{3+} + He \to Li^+ + He^{2+}$, and $B^{5+} + He \to B^{3+} + He^{2+}$ collisions. These excited state helium-like wave functions can be reasonably well described by linear combinations of hydrogen-like bound states within the formalism of the configuration of interactions (CI), as exemplified by, e.g., Bachau [47]. Precisely such excited state wave functions were used for DC in the studies by Gayet *et al.* [22], as well as by Purkait *et al.* [32] and Ghosh *et al.* [23]. It was found in Ref. [22] that excited states were not very important within CDW-4B for DC in He^{2+}-He collisions because of the dominant contribution from the resonant $1s^2 \to 1s^2$ transition. As mentioned, CDW-4B1 is not fully satisfactory for He^{2+}-He collisions, but the situation is conditionally improved by CDW-4B2. Further computations within CDW-4B2 are necessary for this collision to include the ignored off-shell matrix elements and the intermediate as well as final excited states. Intermediate excited states would check the convergence properties of the expansion of the two-center Green's function. Final excited states are important since the transition $He(1s^2) \to Li^+(1s^2)$ in process (2) does not provide the dominant contribution. This is clearly seen in Figs. 1 and 3 where the excited states of Li^+ included within CDW-4B and CDW-IPM, respectively, give a large contribution relative to the cross sections for the ground-to-ground state transition. In particular, Fig. 3 is a good illustration of the caution which one must exercise when comparing theory and experiment, since good agreement found for the ground-to-ground state transition in process (2) disappears altogether when the excited states are taken into account.

B. Li^{3+}+He case

For DC in Li^{3+}-He^0 and B^{5+}-He collisions, especially singly-excited states $(1s,nl)$ with $n \leq 3(l \leq n-1)$ were found to provide significant contributions [22]. At the same time, higher-doubly-excited stated were shown to give cross sections that are at least an order of magnitude smaller than those due to singly-excited states [22]. Overall, despite the inclusion of a restricted set of helium-like excited states, agreement between CDW-4B [22] and experimental data were still only qualitative. This could be due to the replacement of the original CI orbitals $(1s,3l)$ with the actual nuclear charges [47] by the single-configuration product of two hydrogen-like wave functions with certain effective (screened) nuclear charges [22]. Such a simplification, which was motivated by a reduction in computational demands, leads to nonorthogonality between the single-configuration orbitals and the remaining CI orbitals from Ref. [47]. It is known that the excited state wave functions of Bachau [47] are not well adapted to describe the singly-excited states above the $(1s,2l)$ level and, moreover, the modification from Ref. [22] is nonunique. Since helium-like singly-excited states are important for DC, they must be very accurate and should be included by using some other wave functions to verify the results obtained with the CI orbitals from Ref. [47]. It remains to be seen whether this could improve the overall standing of CDW-4B. A more favorable situation was encountered when comparing BCCIS [23] with measurements for He^{2+}-He collisions, despite the same ambiguous modification [22] of the CI wave functions from Ref. [47]. However, for Li^{3+}-He^0 collisions, BCCIS-4B underestimates our measured data similarly to CDW-4B, as seen in Fig. 1.

Regarding the wave functions for the $1s^2$ ground state of helium target, it was found by Belkić and Mančev [34] that total cross sections for DC are weakly sensitive to the number of terms in the CI expansion. Specifically, quite simple 1–4 parameter orbitals of Hylleraas [48], Löwdin [49], Green *et al.* [50], and Silverman *et al.* [51] were shown to be sufficient [34]. Following this finding, Gayet *et al.* [20,22] employed the four-parameter wave function of Löwdin [49] for the initial ground state of helium. Earlier, within IA2-IPM, Gravielle and Miraglia [26] also used a relatively simple Hartree-Fock wave function for $He(1s^2)$ in the form of the Slater-type orbitals given by Clementi and Roetti [52]. This type of simple two-electron wave functions was used not only in CDW, but also in CDW-EIS [20,21] and BCCIS [23,32].

The most unanticipated conclusion from these previous investigations was that CDW-EIS-4B underestimates the experimental data on DC in He^{2+}-He collisions by orders of magnitude at intermediate and MeV/amu energies [41]. This continues to be the case for Li^{3+}-He^0 collisions at high impact energies 2–5.5 MeV from the present measurement, as evidenced by Fig. 1. Such an occurrence is very surprising since at these impact energies, CDW-EIS-3B is known to be remarkably successful. Thus, the above-mentioned conjecture that the number of captured electrons should not change the lower validity limit of CDW does not apply to CDW-EIS. This new unfavorable situation with CDW-EIS-4B casts serious doubts on the whole concept of eikonal electronic continuum intermediate states, since such states for two active electrons totally fail to describe DC at intermediate and high energies. This experience might indicate that with every additional electron to be captured from a multielectron target, the validity of CDW-EIS could be pushed to ever higher energies and this, in turn, would severely limit the usefulness of the eikonal Coulomb multiple electronic states. As stated, in order to see whether the two-step collisional mechanism could improve the prospect for CDW-EIS-4B, Martinez *et al.* [41] took into account the second-order in a perturbation expansion as in IA2-IPM [26]. However, such an attempt has not rescued the situation since this time the resulting cross sections computed by means of CDW-EIS-4B2 were much larger than the experimental data for DC in He^{2+}-He collisions at MeV energies. Thus, CDW-EIS definitely appears as inadequate for DC, as pointed out earlier in Refs. [1,2].

As to CB1-4B for DC in the mentioned pure four-body collisional systems, all the previous computations were carried out only for the ground-to-ground state capture using the one-parameter Hylleraas [48] wave function with the Slater-screened nuclear charge for the initial and final states [24,25]. The obtained total cross sections for H^+-He collisions largely overestimate measurements at all energies [2]. On the other hand, this model agrees well with experiments on DC in He^{2+}-He and Li^{3+}-He^0 collisions at intermediate energies. However, due to the important, but neglected contributions from excited states, such an agreement between CB1-4B and measurements at intermediate energies seen in Fig. 1 for Li^{3+}-He^0 collisions is fortuitous. Thus, it follows that CB1-4B is inadequate for DC [1,2]. This is expected, given that DC is a process with two active electrons for which a first-order model is unsuitable due to the exclusive reliance only upon the direct separate interactions of the projectile with the two target electrons and upon the inclusion of the asymptotic eikonal relative motions of nuclei in the fields of screened nuclear charges in the entrance and exit channels. In other words, one reason for the failure of CB1-4B could be the neglect of continuum intermediate states of the two electrons. This is indeed true, as was demonstrated by Belkić [35,43] in BDW-4B and BCIS-4B that both include the twofold electronic continua in one channel, while in the other channel the descriptions coincide with those from CB1-4B. Computations show that the total cross sections from BDW-4B and BCIS-4B [36,43] are in good agreement with measurements on DC in He^{2+}-He collisions at higher MeV energies where CB1-4B overestimated the experimental data by orders of magnitude. Overall, despite its demonstrated inadequacy for DC, it is still convenient to have the cross sections from CB1-4B to highlight the potential significance of a missing effect due to double continuum intermediate states of the electrons. When CB1-4B is amended by the inclusion of these lacking Coulomb states, BCIS-4B emerges showing reasonable agreement with experimental data for He^{2+}-He collisions [43]. Likewise, in the case Li^{3+}-He^0 collisions, Fig. 1 shows that BCCIS-4B [23] is in a fair agreement with our data at $E \leq 4$ MeV.

Having discussed the salient aspects of the status of theory for two-electron transfer, we can now revisit Figs. 1–3 with a special focus on our experimental data at impact energies larger than those considered in the related previous measurements on DC in Li^{3+}-He^0 collisions. We saw in Fig. 2 that at impact energies $2 \leq E \leq 5.5$ MeV, the present experimental data are reproduced excellently by CDW-EIS-IPM [27]. However, at the same time, our measured total cross sections are grossly underestimated by CDW-EIS-4B, as seen in Fig. 1. At energies $E \geq 2$ MeV, there is also a seemingly good agreement between our experimental data and the predictions by IA2-IPM. However, this is fortuitous due to the neglect of the non-negligible contribution from IA1-IPM (as judged by reference to CIS-IPM in Fig. 2) and because of the ignored intermediate and final excited states of Li^+. The key importance of the final excited states of Li^+ in process (2) is exemplified in Fig. 3 within CDW-IPM. As discussed, it is seen in this figure that a good agreement between CDW-IPM and experiment without the final excited states ceases to exist when the allowance is made for the missing excited states of Li^+. A similar worsening of the apparent agreement between IA2-IPM and our measured cross sections seen in Fig. 2 is anticipated to occur by including the final excited states of Li^+ in the computations.

It can be concluded from the above comparisons between experiment and theory on DC that the situation is not clear cut regarding consistency of performance of the various models. Therefore, more work is needed to better clarify the status of theory for two-electron transfer at high energies. All the existing theoretical models are restricted to sequential capture without any dynamic interelectron correlations. Since static correlations in the bound helium-like systems were not found to be essential for DC, it would be important to clarify the role of dynamic collisional correlations between two electrons. This has not been done thus far. It would also be of interest to examine the contributions from other mechanisms for DC, such as the coherence effect of the simultaneous capture of two electrons. Admittedly, it is very difficult to design a practical theory which would incorporate all these mechanisms and effects for DC, but it is certainly a goal which would be worthwhile to achieve in the near future. From the experimental view point, it would be significant to complement the present data on total cross sections for DC by measuring angular distributions of scattered projectiles at sufficiently high energies. Here, the main goal would be to clearly detect the triple billiard-type collisions with the emergence of the three Thomas peaks predicted theoretically by Belkić *et al.* [36] within the 4B treatments, as opposed to only two such maxima stemming from IPM used by Martínez *et al.* [40]. One of the triple Thomas peaks from the 4B formalism involves explicitly the dynamic electron-electron correlation effect and this is precisely the maximum which is missing from IPM. As such, differential cross sections measured at sufficiently high impact energies would offer an advantageous testing ground for a stringent validation of 4B theories versus IPM on the level of angular distributions, especially when the corresponding total cross sections cannot discriminate between these two approaches.

IV. CONCLUSION

We have measured total cross sections for single capture (SC), double capture (DC), single ionization (SI), double ionization (DI), and transfer ionization (TI) in collisions of Li^{3+} projectiles with He targets at high impact energies $E=2.0-5.5$ MeV. Invariably, all our findings connect smoothly to the corresponding experimental data from earlier measurements at an overlapping intermediate and lower energy range 0.3–2.5 MeV. This ensuing excellent agreement with the past measurements serves as a check of consistency, which is important particularly regarding our independent normalization procedure. In the literature, experimental data for SC, SI, DI, and TI in the investigated collisional system were analyzed in sufficient details and, therefore, we contented ourselves to simply report our related data without a further elaboration on interpretation.

As to DC, however, the status of the existing theories versus experiments is not clear cut. Thus, it was deemed necessary to perform a thorough interpretation and a critical evaluation of the corresponding theoretical methods in the light of the present measurements in concert with the previous experimental data. The two major strategies from the literature on DC, the semiclassical independent particle model (IPM) and the quantum-mechanical four-body (4B) formalism are found to exhibit a varying degree of success when passing from one concrete method to another within which the said strategies were implemented. Certainly, the most frequently used theoretical methodologies with correct boundary conditions are from the realm of the continuum distorted waves (CDW) and continuum distorted wave eikonal initial states (CDW-EIS) in their respective IPM and 4B versions as acronymed by CDW-IPM, CDW-4B, CDW-EIS-IPM, and CDW-EIS-4B. As known, CDW employs full Coulomb waves for initial and final continuum intermediate states for electronic motions in the field of the projectile and target nuclei. These Coulomb wave functions are simplified in CDW-EIS in the entrance channel by means of the corresponding spatially asymptotic logarithmic eikonal phase factors for the active electrons in the projectile field in the initial state. This simplification was initially introduced in the case of SI and SC for the reason of having normalized total scattering wave functions for the initial state of the whole system. Such a simplifying approximation of CDW proved fruitful in practice, since CDW-EIS was systematically found to produce the needed bending of the curves for total cross sections in SI and SC near and below the Massey peak (i.e., in the resonant energy region where the incident velocity matches the classical Bohr velocity in the target orbit from which one active electron is ionized or captured). By contrast, total cross sections in CDW for SI and SC keep on rising with decreased impact energies and this is due to an overabundant density of the continuum intermediate states manifested by the presence of the Coulomb normalization factors. These latter factors disappear altogether from the asymptotic forms of Coulomb waves, or equivalently, from the eikonal initial states, and it is precisely for this reason of a drastically reduced density of continuum intermediate states in SI and SC that CDW-EIS is capable of producing the Massey peaks in accordance with the adiabatic hypothesis. Such a reduction happens to be in the right direction for SI and SC, thus yielding systematically excellent agreements of CDW-EIS with the corresponding experimental data virtually at all energies. This is the case even at lower impact energies that lie far below the expected limit of validity of applicability of first-order terms from perturbation expansion of transition amplitudes.

However, this favorable eikonalization of electronic Coulomb waves for SI and SC turns out to fail completely in CDW-EIS-4B for DC. Specifically, CDW-EIS-4B underestimates our experimental data by a factor which attains even two orders of magnitude and this discrepancy enlarges with decreased impact energy. This indicates that the reduction in the continuum intermediate state density for both electrons in the entrance channel is way too drastic in CDW-EIS-4B to represent a physically adequate motivation for eikonalization of two Coulomb waves. Such a conclusion is supported by CDW-4B, which is in a relatively close proximity of our experimental data. Although, CDW-4B still underestimates our experimental data, it does so by a reasonable, much smaller factor than the dramatic two orders of magnitude in the case of CDW-EIS.

Surprises, however, do not stop here for DC. Thus, when passing onto IPM, all our experimental data on DC are excellently reproduced by CDW-EIS-IPM, but simultaneously overestimated within a factor of 2 or so by CDW-IPM when the final ground and excited states are included. However, this unexpected situation is more an exception than a rule since no systematics exist in outperformance of 4B methods by their IPM counterparts. Rather, the following argument holds true: the relative performance of 4B methods and IPM depends on scattering aggregates. Thus, for some other collisional systems (proton-helium double capture is the clearest example due to the lack of the excited states of the negative hydrogen ion), CDW-4B compares much more favorably to experimental data than does CDW-IPM.

In general, it is well-established in the literature that IPM is inadequate for multielectron transitions. The utility of IPM for DC is mainly in providing a hint on the importance of final excited state contributions via a more manageable computational demands than those required by 4B methods. Following such hints, it was subsequently confirmed also in 4B methods that excited states of Li^{+} for DC in Li^{3+}-$He^{0}(1s^{2})$ collisions indeed play a very important role. A limited set of these excited states was included in the computations within CDW-4B using certain restricted hydrogenic configuration interaction wave functions, but this account is probably too small and insufficiently accurate. Better helium-like wave functions for excited states of Li^{+} should be used in future computations for DC in Li^{3+}-$He^{0}(1s^{2})$ collisions to clarify the situation with CDW-4B.

It is anticipated that CDW-4B within its first order in the perturbation series expansion should be capable of containing the main physics for DC. Should this prove insufficient, the second order in the perturbation expansion could be the next step of investigation. This was already tried in the 4B treatment of DC in He^{2+}-$He(1s^{2})$ collisions with a conditional success within the continuum distorted wave, but proved inadequate for the continuum distorted wave eikonal initial state approximations. These attempts followed the initial second-order two-step mechanism proposed in the IPM version of the impulse approximation with incorrect boundary conditions. Although this latter method is in reasonable agreement with the present experimental data, this might be only apparent and, as such, could cease to exist by allowance of several neglected factors that were not proved to be negligible: (i) the first-order term in the impulse approximation, (ii) the off-shell matrix elements in the Green's function for two-center intermediate states, and (iii) intermediate and final excited states in the exit channel.

Overall, DC continues to stand out as a great challenge to theoretical descriptions. To date, no theory is able to quantitatively and systematically explain the corresponding experimental data for all the collisional systems for which the measurements were performed. Surprisingly, striking discrepancies exist between the supposedly leading 4B methods and IPM when they are confronted with each other even at the highest impact energies in the MeV range at which our measurement was carried out. We hope that the present high-energy experimental data, which are believed to advantageously complement the earlier measured total cross sections at intermediate and low energies, will provide an additional motivation for further progress in theoretical modeling of DC via its different pathways such as sequential and simultaneous mechanisms. Our reported cross sections for the other investigated collisional channels, including SC, SI, DI, and TI, also fill in the lacunae in the high-energy part of the corresponding databases that are important in various applications for modeling of energy losses of heavy particles traversing matter.

ACKNOWLEDGMENTS

This work was partially supported by the Brazilian agencies CNPq/CT-Energ, FUJB, FAPERJ, FINEP, CAPES, and the Rede Nacional de Fusão/Brasil.

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