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SINGLE ELECTRON CAPTURE FROM ALKALI ATOMS INTO SLOW DOUBLY CHARGED IONS-
TRANSFER PROJECTILE EXCITATION

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Abstract - Charge exchange in inelastic collisions of slow (0.5 - 10 keV) doubly charged ions (He$^{2+}$, Ne$^{2+}$, Ar$^{2+}$) with alkali atoms (Li, Na, K) is almost exclusively due to single electron capture (SEC). Since these SEC reactions populate excited projectile states only, they are typical for inelastic multicharged ion – atom collisions. Whereas for He$^{2+}$ the SEC is not accompanied by additional electronic transitions, for Ne$^{2+}$ and Ar$^{2+}$ also final states with rearranged primary ion core states are produced, for which processes a possible involvement of correlated two-electron transitions is of interest. We have applied translational energy spectroscopy to study such SEC processes in detail and present a general account of our results.

1. Introduction
In recent years the principal features of electron capture in collisions of singly and multiply charged ions with neutral atoms/molecules have become increasingly well understood. Quantum-mechanical, semiempirical and classical calculations have been successfully compared to measured total as well as state-selective single electron capture (SEC) cross sections /1/. This understanding, however, becomes rapidly less complete if more-electron transitions related to electron capture are considered, e.g. double electron capture, transfer ionisation or transfer excitation of projectile or target. For collisions of slow Ne$^{2+}$ or Ar$^{2+}$ with Li(2s), SEC is much more important than double electron capture. It can take place with or without conservation of the initial projectile ion state, as we have shown in a recent paper /2/. The “core-violation” processes (transfer projectile excitation or deexcitation) involve transitions of at least two electrons in a single SEC event. We have extended these studies, carried out by means of translational energy spectroscopy, in a systematic way to Na(3s) and K(4s) target atoms. Moreover, for Ar$^{2+}$ primary ions the population of highly-excited metastable Ar$^+$ states has been investigated to facilitate the interpretation of reaction channels.

In the following we discuss typical results on core-conserving (“CC-SEC”) as well as core-violating (“CV-SEC”) SEC reactions, which have been studied with Ne$^{2+}$ and Ar$^{2+}$ primary ion beams of different composition and kinetic energy.
2. Experimental Method
We used a translational energy spectrometer as shown in fig. 1, basically consisting of a Nier-type ion source, an analyzer magnet, a collision chamber with an effusive alkali atom beam, a deceleration-energy analysis stage for both primary and charge-exchanged projectile ions, and an ion detector unit. The last one (cf. insert in fig. 1) has been used in two detection modes. Ions leaving the analyzer are post-accelerated up to 2.5 keV and will be detected almost irrespective of their internal energy. If, however, ions drift into the CEM detector just with the pass energy of the analyzer (20 eV), only highly excited species are counted. Under the given circumstances these are metastable ions, which for the present collision systems can only be produced from the Ar\(^{2+}\) projectiles. The obtained translational energy spectra ("TES") show an energy resolution of 0.4 eV FWHM as demonstrated in fig. 2 for SEC in He\(^{2+}\)-Li collisions.

The electron impact energy of the Nier-type ion source could be adjusted to control the \(^1D\) and \(^1S\) metastable admixtures in Ne\(^{2+}\) or Ar\(^{2+}\) primary ion beams including their complete suppression /3/. The scattering angle was variable around forward direction by up to \(\pm 3^\circ\), to check the impact-parameter dependence of TES. A more complete description of apparatus and technique of metastable ion detection will be given elsewhere (Schweinzer et al., in preparation).

3. Discussion of Results
We measured TES for impact of He\(^{2+}\), Ne\(^{2+}\) and Ar\(^{2+}\) on Li, Na and K and also on some other metal targets as Mg and Hg. In this paper we comment only on a few collision systems, to present essential findings of these investigations.
3.1 He\textsuperscript{2+}-Li

Fig. 2 shows a TES for SEC in collisions of 10 keV He\textsuperscript{2+} with Li, for which the population of He\textsuperscript{+}(n=3) is dominant, but capture into He\textsuperscript{+}(n=2,4,5)-states is clearly visible, too. In an earlier study\textsuperscript{44} we have already presented TES for this collision system together with references to related work from other groups, covering measured as well as calculated total and state-selective SEC cross sections.

![Fig. 2: TES for SEC in He\textsuperscript{2+} - Li(2s) collisions at 10 keV impact energy. Arrows indicate He\textsuperscript{+} principal shells populated by SEC.](image)

The here presented TES are of higher quality than our earlier ones, however. Combination with data from photon-spectroscopical investigations (Aumayr et al., in preparation) permits a rather detailed comparison with elaborate quantum-mechanical calculations from different groups on this quasi-one-electron collision system. The experimental data are also of interest for lithium beam-activated charge exchange spectroscopy in tokamak plasmas\textsuperscript{55}.

3.2 Ne\textsuperscript{2+}-Li

This collision system is especially interesting considering correlated two-electron transitions related to SEC. Fig. 3a shows the relevant final states Ne\textsuperscript{+}(n,l) produced via SEC with corresponding reaction energy defects. As already explained in detail in\textsuperscript{22}, in this system CC-SEC as well as CV-SEC are observed with the latter becoming dominant toward low impact energy (cf. fig. 3b). CV-SEC involves capture into Ne\textsuperscript{2+} \textsuperscript{1D} primary ions leading to Ne\textsuperscript{+} final states with Ne\textsuperscript{2+} \textsuperscript{3P} core (transfer projectile deexcitation, channels 3,5,6) as well as capture by Ne\textsuperscript{2+} \textsuperscript{3P} into Ne\textsuperscript{+} final states with Ne\textsuperscript{2+} \textsuperscript{1D} core (transfer projectile excitation, channel 4), respectively.
Fig. 3a: Final state energy levels relevant for SEC in Ne$^{2+}$ - Li(2s) collisions. Chequered fields contain levels populated via CV-SEC. Arrows indicate TES peaks in fig. 3b.

Fig. 3b: TES for SEC in Ne$^{2+}$ - Li(2s) collisions at different impact energies. Arrows indicate reaction channels discussed in text.

This assignments could be unambiguously proven by varying the Ne$^{2+}$ $^3P_1^o$/$^3D$ ratio as described in chapter 2. The CC-SEC (in first instance channel 2) becomes relatively more important at high impact energy, whereas at lower impact energy the CV-SEC starts to take over. The importance of different reaction channels can be explained with reference to a multichannel Landau-Zener treatment of the SEC processes /1,6/. The resulting so-called reaction window of energy defects $\Delta E$ shows which SEC processes are favoured. In the present case the CV-SEC reaction channels happen to involve more favourable energy defects than the CC-SEC channels. However, because of the smaller coupling strengths for two-electron transitions as compared to one-electron transitions the probability for CV-SEC can only become appreciable at low impact velocity, where the unfavourably situated CC-SEC channels are strongly suppressed (see also discussion on simultaneous transitions in section 3.4).

3.3 Ar$^{2+}$-Li,Na,K
The relevant final state energy levels are shown in fig. 4a for the case of Na, whereas for Li and K a
shift of the ΔE-scale by respectively 0.25 downwards and 0.8 eV upwards is appropriate, considering the ionisation energies of 5.39, 5.14 and 4.34 eV for Li, Na and K, respectively. For these three target species the relative importance of equivalent SEC reaction channels varies dramatically as seen from fig. 4b, where TES have been arranged such that identical final Ar\(^+\)-states are vertically aligned. Whereas for Li CC-SEC (channel 3) is dominant at all impact energies from 0.8 up to 6 keV (cf. /2/), for K CV-SEC (channel 2) dominates completely with CC-SEC (channel 1) keeping up only toward the highest impact energies investigated (12 keV). The case of Na is intermediate with CV-SEC dominating below ca. 1.5 keV impact energy and CC-SEC above. CV-SEC (channel 2) is due to SEC of Ar\(^{2+}\)\(^3\)P into Ar\(^+\) (\(^1\)D)4p;3d final states (transfer projectile excitation). Of particular interest is the population of metastable Ar\(^+\) 3d;3d′ states, which could be directly observed as shown in chapter 2. These metastable states (marked by their LS-terms in fig. 4a) cause the hatched TES displayed in fig. 4b. As will be explained in detail elsewhere (Schweinzer et al., in preparation), for the present collision systems the metastable states are mainly populated via cascade transitions from initially produced Ar\(^+\) 4p;4p′ final states, whereas e.g. in Ar\(^{2+}\)-Mg collisions a direct population of the metastable states takes place. This difference causes a much higher metastable fraction for Ar\(^+\) ion beams produced in Ar\(^{2+}\)-Mg collisions than with the alkali metal targets.

3.4 Hints to Correlated Two-Electron Transitions
In recent studies on more-electron transitions in ion-atom collisions the involvement of electron-electron correlation has attracted considerable interest and some debate /7,8/. We consider potential energy curves for the relevant collisional quasi-molecular states. All SEC exit channels exhibit Coulomb repulsion and consequently cross the potential energy curve of the entrance channel at internuclear distances \(R/a_0=27.2/\Delta E(\text{eV})\). From our previous work /2/ we already know, that the reaction window for SEC from Li into doubly charged ions favours energy defects of \(2 \leq \Delta E \leq 3 \text{ eV}\). Approximately the same can be concluded for SEC from Na and K, cf. fig. 4b. In the case of Ne\(^{2+}\)-Li, the dominant CV-SEC channels involve \(R_e\)-values of about 20 \(a_0\) for (3) and (4) and about 10 \(a_0\) for (5), whereas the CC-SEC channels cross at about 30 \(a_0\) for (2) and at about 6 \(a_0\) for (7), respectively, cf. /2/. Particularly at low impact energy a two-step population of (5), involving as a first step CC-SEC into (2) or (7) can be ruled out, which strongly suggests a simultaneous two-electron transition for CV-SEC into (5). In a similar way, in Ar\(^{2+}\)-K collisions the CV-SEC (channel 2) is assigned to a simultaneous two-electron transition, whereas for Ar\(^{2+}\)-Na and Ar\(^{2+}\)-Li its population in two steps via the CC-SEC channel (3) cannot be excluded, as was deduced from the TES-scattering angle dependence.

From the size of presently studied total SEC cross sections we are able to derive the absolute magnitude of various coupling strengths governing the above discussed reactions. This information will permit a decision on whether the above identified simultaneous two-electron (CV-SEC) transitions are of correlated or uncorrelated nature /7,8/.
**Fig. 4a:** Final state energy levels relevant for SEC in Ar$^{2+}$ - Na(3s) collisions. Chequered fields contain levels populated via CV-SEC. Arrows indicate TES peaks in fig. 4b. LS-terms indicate metastable Ar$^+$ final states.

**Fig. 4b:** TES for SEC in Ar$^{2+}$ - Li(2s), Na(3s), K(4s) collisions at 2 keV impact energy. Hatched TES refer to metastable states only.

**Conclusions**
For Ne$^{2+}$ or Ar$^{2+}$ colliding with Li, Na or K, as a general result single electron capture with or without change of the primary ion core states is mainly governed by the situation of involved reaction energy defects with respect to the reaction window. Two-electron transitions in core-violating SEC become relatively more probable toward lower impact energy. From the corresponding total SEC cross sections coupling strengths for relevant SEC transitions can be estimated, which permit quantitative conclusions on the involvement of correlated two-electron transitions.

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