Multielectron removal processes in He^{2+}+Na collisions

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Multielectron processes have been studied by measuring the Na^{2+} and Na^{3+} recoil momenta resulting from 10 keV/amu He^{2+}+Na(3*s*) collisions. The Na^{2+} *Q*-value spectrum shows that transfer ionization dominates two-electron removal. Double capture populates mostly singly excited He(1*snl*) states. A smaller fraction of double capture leads to doubly excited He. Na^{3+} recoil ions are created by double capture into the He ground state and the emission of a third electron into the continuum. The Na^{3+} recoil ion is not left in its triplet ground state but in one of the low-lying excited singlet terms due to spin conservation.

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I. INTRODUCTION

Electron-capture processes in keV collisions of highly charged ions with various atomic and molecular targets play an important role in fusion and astrophysical plasmas. The understanding and theoretical modeling of one-electron capture from (quasi-)one-electron targets (alkali-metal atoms and atomic hydrogen) is rather well established [1,2], but the knowledge of multielectron capture processes is much less developed. However, these processes can play a significant role [3–5], e.g., in solar-wind-induced cometary x-ray emission. Many experiments using rare-gas targets showed that a multitude of processes determines the charge balance [6–12]. For example, charge states of Ar target ions higher than the initial projectile charge state are observed for C^{6+}, N^{6+}, and O^{6+}, implying that even in low-keV collisions of highly charged ions on gas targets excitation of the target occurs due to inner-shell electron capture [7]. Studies of multielectron capture involving alkali-metal targets are rather scarce. For alkali-metal targets the relative importance of multielectron processes compared to one-electron capture is smaller, because of the low binding energy of the valence electron. However, the absolute cross sections of multielectron processes are in the same range as the ones for rare-gas targets (see, e.g., [13]).

We will address multielectron removal in 10 keV/amu He^{2+}+Na collisions by measuring the momenta of the Na^{2+} and Na^{3+} recoil ions using the technique of magneto-optical trapping recoil-ion-momentum spectroscopy (MOTRIMS) [14–16]. For this collision system absolute cross sections for multielectron processes have also been determined by means of the growth-curve method [17] and coincidence techniques [13]. As already noticed by DuBois [13], an apparent inconsistency showed up between these pioneering experiments, as true double capture (σ_2^{20}) was found to be larger than the total He^0 production ($\sigma^{20}=\sigma_2^{20}+\sigma_3^{20}+\cdots$) by a factor of 2. The present MOTRIMS study will clarify this issue.

Our previous MOTRIMS measurement showed that Na^{3+} recoils are created by a combined process of double capture into the He ground state and single ionization [18]. An interesting issue is the final state of Na^{3+} recoil ions after three-electron removal. It was argued that spin blocking might prohibit population of the energetically most favorable Na^{3+} state, the triplet ground state. However, the triplet and nearby lying singlet terms could not be resolved. Due to the improved resolution of our experiment the present MOTRIMS study allows the role of spin conservation to be addressed.

II. EXPERIMENT

Our MOTRIMS apparatus has been described elsewhere [18,19]. In short, sodium atoms are cooled and trapped in a magneto-optical trap (MOT) which is crossed by a chopped ion beam. The resulting Na^{r+} recoil ions are extracted in the transverse direction by means of a weak electric field toward the detector where both the two-dimensional position and the time of flight are recorded. From this data the recoil momentum vector can be reconstructed. Note that the recoil spectra reveal the primary population, thus before the projectile decays by photon or Auger electron emission. The laser beams of the MOT are switched off in synchronization with the ion beam pulse, such that during the collisions no laser light is present and all Na atoms have decayed to the ground state.

The different recoil charge states are distinguished by means of their time of flight (see Fig. 1). Typically for collisions between multiply charged ions and Na the amount of Na^{2+} is only a few percent of Na^+, due to the large difference in ionization potential between Na ($I_1=5.14$ eV) and Na^+ ($I_2=47.3$ eV). By integrating the Na^{r+} peaks one obtains relative cross sections for one- or more-electron removal. From well-known absolute experimental data for one-electron capture [17,20] and recently obtained single-ionization cross sections [21] the Na^+ cross section can be put on an absolute scale and thus also the Na^{2+} and Na^{3+} ones.

III. RESULTS AND DISCUSSION

A. Two-electron removal

The longitudinal component of the Na^{2+} recoil momentum after double capture (DC) is related to the *Q* value, i.e., the difference between the total binding energies before and after the reaction, via the relation (in atomic units) [22]

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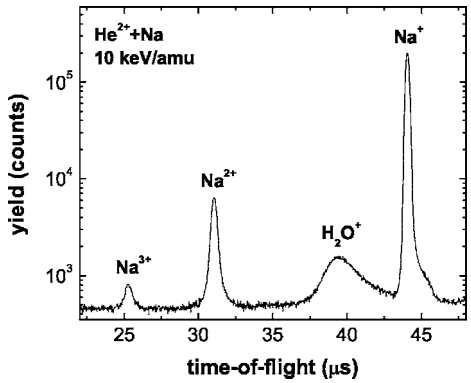

FIG. 1. A time-of-flight spectrum of recoil ions resulting from 10 keV/amu He^{2+}+Na(3*s*) collisions. Note that the presence of background H_2O^+ recoils is enhanced because they are formed along the whole ion beam path (~5 cm), while the Na target has a diameter of about 1 mm only.

$$p_{long} = \frac{Q}{v_p} - v_p, \tag{1}$$

with v_p the velocity of the projectile ion and $Q = I_1 + I_2 - E_b^{\mathrm{He}}$, where E_b^{He} is the total binding energy of the final He state (positively defined). Na^{2+} recoils can also be created by transfer ionization (TI), whereby one electron is transferred to the projectile and one is emitted into the continuum. The corresponding longitudinal momenta are given by

$$p_{long} > \frac{I_1 + I_2 - E_b^{\mathrm{He}^+}}{v_p} - v_p, \tag{2}$$

where $E_b^{\mathrm{He}^+}$ is the binding energy of the final He^+ state. When converting longitudinal momentum into a Q value using Eq. (1) TI is expected at $Q > I_1 + I_2 - E_b^{\mathrm{He}^+}$. TI leading to $He^+(1s)$ would appear at $Q > -2.0$ eV and $He^+(n=2)$ at $Q > 38.8$ eV. Double ionization (DI) would lead to longitudinal momenta of

$$p_{long} > \frac{I_1 + I_2}{v_p} - v_p, \tag{3}$$

which in the Q-value spectrum correspond to a $Q > I_1 + I_2 = 52.4$ eV.

The Q-value spectrum of the Na^{2+} recoil ions is shown in Fig. 2. The momentum resolution of the Na^{2+} recoils was 0.18 a.u., corresponding to a Q-value resolution of 3 eV. The largest contribution is found around $Q \sim 0$ eV and originates from TI into the $He^+(1s)$ state and DC into He($1snl$, $n \geq 2$). At larger Q values a small amount of DC into doubly excited He states appears. DC into He($2lnl'$) and higher excited states can be distinguished. From the spectrum it is seen that DC into the symmetric He($2l2l'$) states is more likely than into He($2lnl'$, $n \geq 3$). The spectrum decreases sharply at the boundary of DI, suggesting that this process is highly unlikely to occur. No contribution from DC into the He($1s^2$) ground state is found. Furthermore the minimum around $Q \sim 40$ eVsuggests that TI with capture into $He^+(n=2)$ is at best weak.

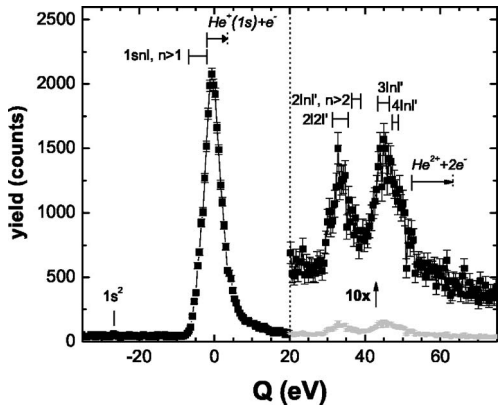

FIG. 2. Q-value spectrum of Na^{2+} recoils. The relevant He states are indicated, as well as the boundaries to transfer ionization (−2.0 eV) and double ionization (52.4 eV). The arrows show the Q-value ranges in which transfer ionization or double ionization can be expected.

Our obtained cross sections are listed in Table I. The method to obtain separate cross sections for TI and DC populating He($1snl$, $n \geq 2$) is similar to that used for the separation of one-electron capture and single ionization [21,23]. Because in the present work the final projectile charge states are not measured a direct comparison with the coincidence measurements is not always possible. In the coincidence measurements the flight time from the collision center to the projectile detector was long compared to the lifetime of the excited projectile states [13]. Therefore the σ_2^{21} cross section contains besides TI also DC to states that deexcite by autoionization, autoionizing double capture (ADC). Only DC to states that decay via photon emission, true double capture (TDC), contributes to σ_2^{20}. Singly excited He($1snl$, $n \geq 2$) states can only decay radiatively and therefore contribute to

TABLE I. Absolute cross sections for the different two- and three-electron removal processes resulting from He^{2+}+Na(3*s*) collisions at 10 keV/amu. The errors include a systematic uncertainty of 15–20 % due to our normalization procedure. In the nomenclature σ_j^{2i} the indices i and j are the final charge states of the projectile and the target, respectively, while σ_j represent the cross sections for the total Na^{j+} recoil-ion production.

Process	Cross section (10^{-16} cm^2)	
σ_2	5.2±0.8	
σ_3	0.4±0.1	
TI	3.7±0.6	
DC He($1snl$, $n \geq 2$)	0.9±0.3	
DC He($nln'l'$, $n \geq 2$)	0.6±0.2	
σ_2^{20}	1.2±0.5	7.0±3.5[a]
σ_2^{21}	4.0±0.9	3.5±0.7[a]
σ_3^{20}	0.4±0.1	1±0.5[a]
σ_3^{21}		<0.2[a]
$\sigma^{20} = \sigma_2^{20} + \sigma_3^{20} + \cdots$	1.6±0.5	3.1±0.4[b]

[a]DuBois [13].
[b]Dubois and Toburen [17].

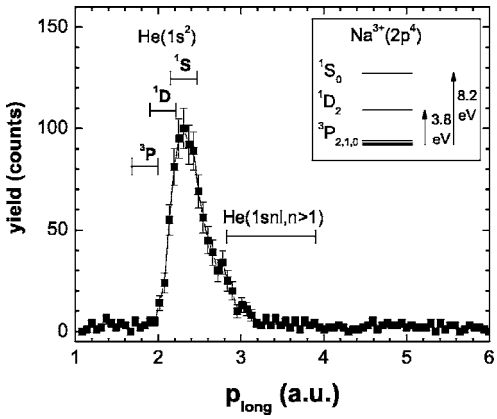

FIG. 3. A longitudinal momentum spectrum of Na^{3+} recoils. The momentum resolution is 0.4 a.u. The assignment of the different target and projectile final states is explained in the text.

TDC. The population of He($nln'l'$, $n \geq 2$) states can contribute to both ADC and TDC. In order to deduce the σ_2^{20} and σ_2^{21} cross sections for simplicity equal sharing of the He($nln'l'$, $n \geq 2$) contribution over ADC and TDC has been assumed. The uncertainty introduced by this procedure is taken into account in the error bars. The present σ_2^{21} is in agreement with DuBois [13], while there is a large discrepancy concerning σ_2^{20}. The present cross section for total He^0 production, σ^{20}, is somewhat lower than the one of DuBois and Toburen [17]; however, the difference is much smaller than in the σ_2^{20} case. This suggests that the apparent inconsistency between σ_2^{20} and σ^{20} is due rather to σ_2^{20} than to σ^{20}.

B. Three-electron removal

A longitudinal momentum spectrum of Na^{3+} recoil ions is shown in Fig. 3. Here a momentum resolution of 0.4 a.u. was obtained. All the Na^{3+} recoils are found fully in the forward direction, indicating energy loss. The longitudinal momenta are too small for triple ionization or double ionization accompanied by single capture. Therefore the combination of double capture and single ionization (DCSI) leads to Na^{3+}. The following process was proposed [18]. Two inner-shell $2p$ electrons are captured while the outer $3s$ electron is emitted. The closest-lying state in the He^{2+} projectile suitable for capturing strongly bound $2p$ electrons is the He ground state, leading to a large positive Q value of $\sim$45 eV. Note that population of any other He state would lead to even higher Q values.

DCSI involves ionization and because the momentum of the emitted electron is not measured one cannot deduce the Q value from the longitudinal momentum unambiguously. However, for a given He final state there is a minimum longitudinal momentum connected to a zero kinetic energy of the emitted electron in the projectile frame, i.e., electron capture into the continuum (ECC), leading to

$$p_{long} \geq \frac{I_1 + I_2 + I_3 - E_b^{\mathrm{He}}}{v_p} - \frac{3}{2} v_p, \tag{4}$$

where I_3=71.6 eV is the third ionization potential of Na. Assuming small transverse momenta of the emitted electron and excluding the possibility of either backward emission or the situation that the electron is faster than the projectile, a maximum longitudinal momentum is connected to electron excitation into the continuum (EEC), in which the electron stays near the target, which gives rise to

$$p_{long} \leq \frac{I_1 + I_2 + I_3 - E_b^{\mathrm{He}}}{v_p} - v_p. \tag{5}$$

Besides the uncertainty associated with the unknown momentum of the emitted electron also the final electronic state of the Na^{3+} recoil is *a priori* not known. The Na^{3+} ground term is a triplet ^3P, while the first two excited terms are of singlet character, i.e., $^1D^2$ and $^1S^0$. They are excited by only 3.82 and 8.24 eV, respectively. For an excited Na^{3+} final state one has to replace I_3 by $I_3 - E_{\mathrm{exc}}^{\mathrm{Na}^{3+}}$ in Eqs. (4) and (5). The relevant combinations of projectile and target states are indicated in Fig. 3. The range of the Na^{3+} final states for the population of He($1s^2$) is determined by the limit values set by ECC and EEC. Also the range of He($1snl$, $n \geq 2$) is indicated, without specifying the final Na^{3+} states.

From the spectra it is clear that the main channel is capture into He($1s^2$), although a small contribution from capture into He($1snl$, $n \geq 2$) cannot be excluded completely. The final target states appear to be of singlet character. The energetically most favorable situation of population of the ^3P ground term seems very unlikely. This may be explained by a spin-conservation argument. Initially the He^{2+}+Na system forms a doublet. As the He ground state and the emitted electron form a doublet, spin conservation dictates that the final Na^{3+} state is of singlet or triplet character. Furthermore, if the He ground state is populated with two $2p$ inner electrons the Na^{3+} core is left as a singlet. Only spin exchange with the Rydberg-like outer electron could lead to the triplet Na^{3+} ground state, but because of the large energetic separation of the $2p$ and $3s$ electrons such an interaction is unlikely.

IV. CONCLUSION

Multielectron processes in 10 keV/amu He^{2+}+Na($3s$) collisions have been studied by measuring the Na^{2+} and Na^{3+} recoil momenta. The Na^{2+} Q-value spectrum showed that transfer ionization dominates two-electron removal. Double capture populates mostly singly excited He($1snl$) states. A smaller fraction of double capture leads to doubly excited He. Within the latter contribution double capture into He($2lnl'$) and higher excited states could be resolved. Furthermore, population of symmetric He($2l2l'$) states seems more probable than that of asymmetric He($2lnl'$, $n \geq 3$) states. Our transfer ionization cross section is in agreement with previous coincidence measurements; however, we find a much smaller cross section for true double capture. This suggests that the apparent inconsistency in the pioneering work of DuBois is due to the σ_2^{20} rather than the σ^{20} measurement. Na^{3+} recoil ions are created by double capture of two inner-shell electrons into the He ground state, while the outer-shell electron is emitted into the continuum. Due to spin conservation the Na^{3+} recoils are not left in the triplet ground state but in one of the nearby singlet excited states.

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