

# Single Electron Capture in Slow Collisions of Doubly Charged Ions with Dinuclear Molecules

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**Abstract:** We have performed translational-spectroscopical measurements on single electron capture (SEC) by impact of slow (impact energy  $\leq 1$  keV) doubly charged ions on dinuclear molecules. For impact of  $C^{2+}$  and  $N^{2+}$  ions the influence of their metastable fractions on the SEC probability has been studied. For impact of  $^4He^{2+}$  on  $O_2$  and CO direct and dissociative SEC have been investigated. For all collision systems studied the principal SEC channels can be explained by the "reaction window" which results from multichannel-Landau-Zener treatments for the collisional quasimolecules.

**Keywords:** Single electron capture, dissociative electron capture, doubly charged ions

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## Introduction

Collisions of slow doubly charged ions with simple molecules are of interest in hot thin plasmas for various fields, in particular ionospheric physics, plasma technology and thermonuclear fusion. As an important example, in the next generation of magnetically confined fusion plasma experiments an important part of the plasma heating will be provided by fast alpha particles ( $He^{2+}$ ) from D-T fusion reactions. In such so-called burning fusion plasmas the  $He^{2+}$  ions will transfer their initial kinetic

energy of 3.5 MeV in series of elastic collisions to the deuterium and tritium fuel ions, and after having slowed-down end up as "He ash". The latter must be efficiently removed from the plasma in order to avoid unacceptable bremsstrahlung loss and fusion fuel dilution. This involves so far not well understood collective plasma processes in the outermost plasma region where atomic collisions with  $\text{He}^{2+}$  are of relevance. In the plasma scrape-off layer (SOL) appreciable molecular concentrations are generated by particle- and radiation-stimulated desorption from the wall components.

More generally, in collisions of doubly charged ions with neutral molecules single electron capture (SEC) is a highly probable process which can produce molecular ions in both bound and unbound (dissociated) states, depending on the given internal energy. In hot thin plasmas doubly charged ions and neutral molecules may coexist. Hence, a satisfactory understanding of the underlying reaction kinetics requires knowledge of cross sections for the direct (i.e. non-dissociative) and the dissociative SEC processes.

In our first study on such processes we have investigated the collision systems  $\text{C}^{2+}$  -  $\text{H}_2$ ,  $\text{N}_2$ ,  $\text{O}_2$  and  $\text{CO}$  for ion impact energies between 0.2 and 1 keV (cf. [1] and references therein) by means of final-vibrational state-resolved translational energy spectroscopy. The obtained translational energy spectra (TES) showed that both  $2s^2\ ^1\text{S}$  ground state- and  $2s2p\ ^3\text{P}^o$  metastable  $\text{C}^{2+}$  ions can give rise to important SEC channels if the corresponding reaction energy defects fall into the so-called "reaction window" (moderately exothermic reactions with about  $3 \leq \Delta E \leq 6$  eV ( $\Delta E$  is the respective kinetic energy gain)). This reaction window describes the range of strong couplings for SEC in the collisional quasimolecules, as can be concluded from corresponding multichannel Landau-Zener calculations [2]. However, in order to know the relative importance of SEC channels starting from different primary ion states, we need their absolute ion beam fractions. We have applied an experimental technique based on preferential attenuation via electron capture, which can be used in conjunction with TES measurements to determine the ground- and metastable ion beam fractions. The method will be demonstrated for the collision systems  $\text{C}^{2+}$  -  $\text{O}_2$  and  $\text{N}^{2+}$  -  $\text{CO}$ . The main emphasis of the present paper is on SEC by impact of  $\leq 1$  keV  $\text{He}^{2+}$  on  $\text{O}_2$  and  $\text{CO}$ , where in contrast to the other projectile ions no problems arise from metastable fractions, but SEC may lead to dissociation of the molecular product ions. Similar studies by means of translational energy spectroscopy have already been performed by other groups for  $\geq 2$  keV  $\text{He}^{2+}$  impact on  $\text{D}_2$  [3],  $\text{N}_2$  [3],  $\text{O}_2$  [3, 4] and  $\text{H}_2$  [5], and below 2 keV impact energy on  $\text{H}_2$  [6],  $\text{N}_2$  [6] and  $\text{CO}$  [6, 7] target molecules.

## Experimental method

Translational energy spectroscopy measures the kinetic energy change of projectile ions which have been (nearly) forward-scattered in the collisions of interest. In this case the projectile receives the inelastic energy loss or gain in the respective SEC reaction channel almost completely [1], and different reaction channels can be identified in the resulting translational energy spectra (TES).

Change of the projectile scattering angle selects different impact parameter ranges and can thus enhance or suppress different SEC channels with respect to each other. Doubly charged primary ions were extracted from a 5 GHz ECR ion source [8], magnetically mass-to-charge analyzed and guided into the translational energy spectrometer (cf. fig. 1).

Electrostatic lenses provide for ion beam deceleration and -focusing and a hemispherical monochromator for kinetic energy definition, followed by the molecular gas target and a hemispherical energy analyzer with channel electron multiplier for counting the charge-exchanged projectile ions. The kinetic energy resolution of  $\leq 200$  meV permits distinction between different final vibrational states for given electronic transitions [1]. By means of a differentially-pumped charge-exchange cell in front of the translational energy spectrometer the primary-ion beam composition can be varied (see below).

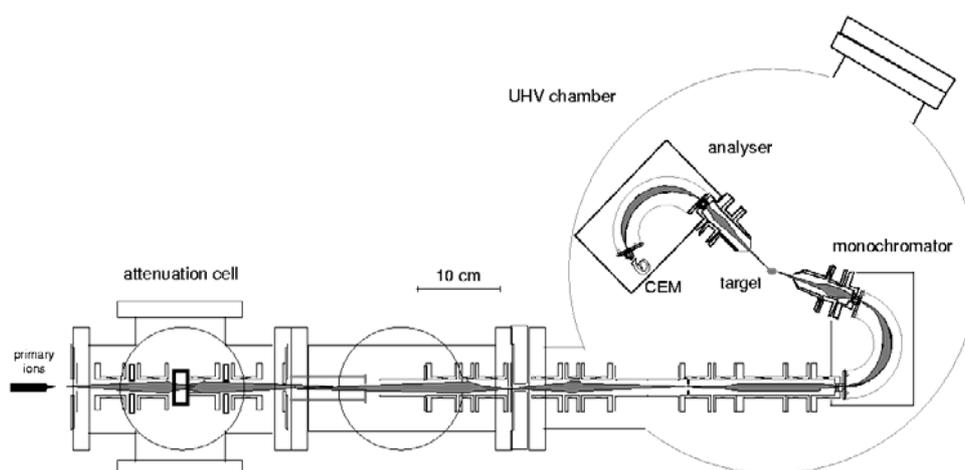


Fig. 1

**Fig. 1** : Schematic view of the translational energy spectrometer with attenuation cell in front.

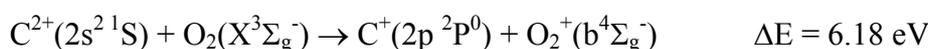
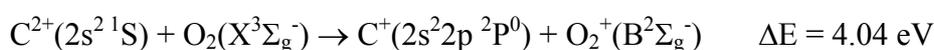
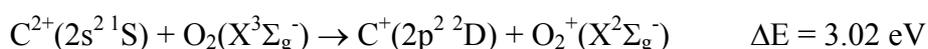
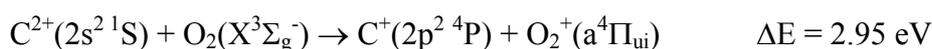
## Discussion of experimental results

### 1. Influence of metastable states in the primary ion beam

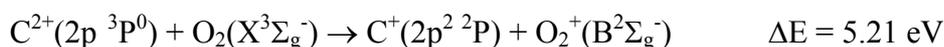
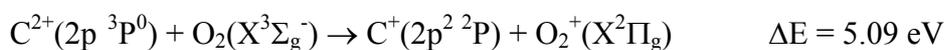
Undefined metastable primary ion beam fractions are a notorious source of experimental uncertainty in low-energy ion collisions, and various techniques may be used as remedy (see [9] and refs. therein). "Double-translational spectroscopy" involves a first translational spectrometer for preparing primary ions via SEC into well defined states, which then are used in a second translational spectrometer for the SEC studies of actual interest [10]. Here we have utilized the technically less

complex "translational attenuation method" [11] for dealing with metastable primary ion beam fractions. To this purpose the attenuation cell (cf. fig. 1) in front of our translational energy spectrometer is filled with an appropriate target gas. When passing the mixed beam of ground state- and metastable ions through this attenuation cell, the two or more ion beam components may undergo SEC with different efficiency, which can be seen from characteristic changes in the subsequently measured TES. For the collision system  $C^{2+} - O_2$ , a well-resolved translational energy spectrum from [1] is shown in fig. 2a.

Four SEC channels have been identified which result from  $C^{2+}(2s^2\ ^1S)$  ground state ions:

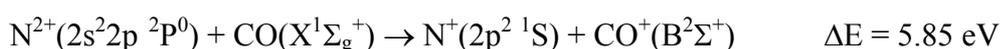
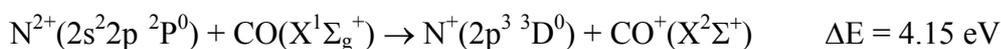


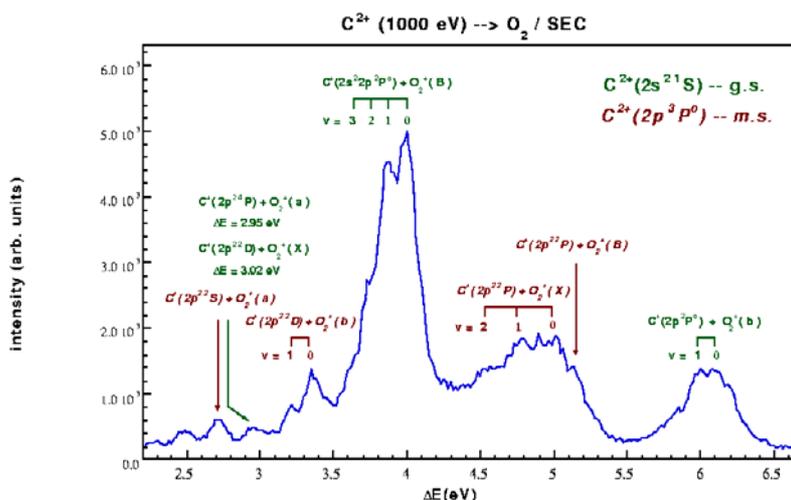
Three other SEC channels belong to the metastable  $C^{2+}(2p\ ^3P^0)$  ion (indicated by italic type):



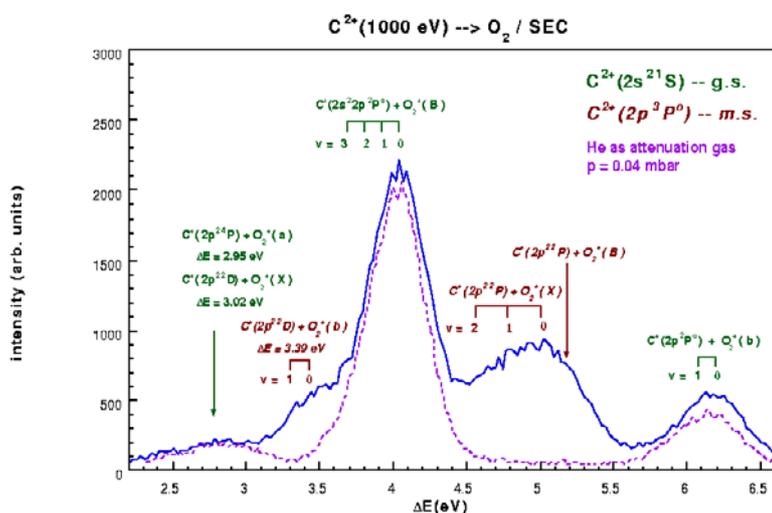
In the above reaction channels, energy gains apply to vibrational ground states of the respective product molecular ions. For ion beam attenuation we have used He in the attenuation cell, keeping in mind that then the reaction energy window favours SEC by metastable  $C^{2+}$  ions [9]. The attenuation gas pressure was 0.04 mbar as measured with an absolutely calibrated capacitance manometer. The consequent change of the primary beam fractions because of the strongly different SEC is clearly apparent from a corresponding change of the low-resolution TES shown in fig. 2b. This supports our earlier interpretation of this somewhat complicated TES [1]. An absolute determination of the metastable ion beam fraction can be made if different attenuation gas pressures are applied [11].

Another example concerns the collision system  $N^{2+} - CO$ . Fig. 3a shows a TES measured for 800 eV impact energy [12]. The two principal peaks are ascribed to SEC into  $N^{2+}(2s^2 2p\ ^2P^0)$  ground state ions:





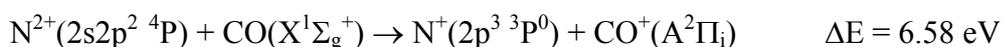
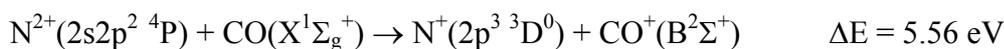
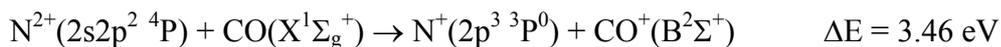
(a)

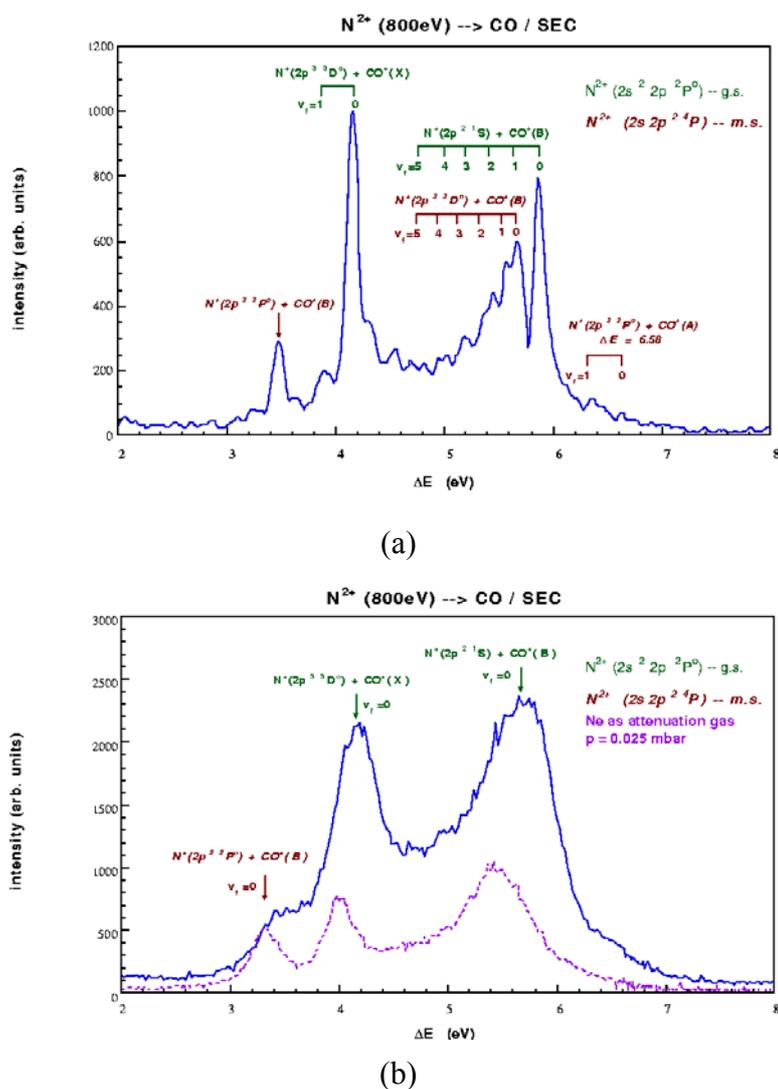


(b)

**Fig. 2:** (a) Translational Energy Spectrum for SEC in collisions of  $C^{2+}$  with  $O_2$  [1]. (b) TES for SEC in collisions of  $C^{2+}$  with  $O_2$  (dotted curve for strong attenuation of metastable  $C^{2+}$  primary ions).

Three minor peaks are assigned to the  $N^{2+}(2s2p^2\ ^4P)$  metastable state ions (indicated by italic type):



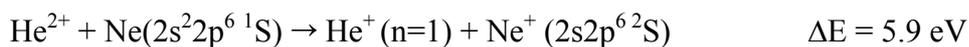


**Fig. 3:** (a) TES for SEC in collisions of  $N^{2+}$  with CO [12]. (b) TES for SEC in collisions of  $N^{2+}$  with CO (dotted curve for strong attenuation of ground state  $N^{2+}$  primary ions).

Fig. 3b shows two low-resolution TES, of which one has been measured with Ne at a pressure of 0.025 mbar in the attenuation cell. This resulted in preferential attenuation of the  $N^{2+}$  ground state fraction as expected from the SEC energy gains for  $N^{2+}$  - Ne collisions, keeping in mind the position of the reaction window.

## 2. SEC by $He^{2+}$ from $O_2$ and CO

At low impact energy the reaction window favours moderately exothermic reactions ( $3 \leq \Delta E \leq 6$  eV) over endothermic or strongly exothermic SEC reactions, but with increasing impact energy such pronounced selectivity gradually disappears [5, 10]. Since TES for  $He^{2+}$  can show some broad structureless features, we have utilized the well defined SEC reaction

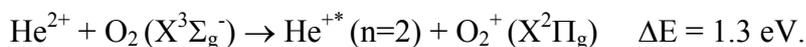


for calibration of the TES kinetic energy gain-/loss scale, by adding Ne to the molecular target gases of interest into the collision cell of our translational energy spectrometer.

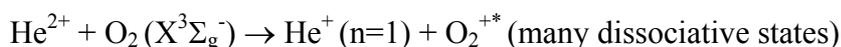
If the fraction of Ne in the target gas mixture and the above state-selective SEC cross section are known, this provides a convenient way for measuring the absolute state-selective SEC cross sections for the molecular target of interest, by comparing the corresponding TES features with each other. The state-selective SEC cross section for Ne can be conveniently measured because at low impact energy this reaction channel is the only one favoured by the reaction window and therefore its cross section agrees with the total SEC cross section. The latter one can be measured in the further course of this work for the impact energy range of interest. First results of our present studies for SEC by  $\text{He}^{2+}$  are summarized as follows.

### $\text{He}^{2+} + \text{O}_2 / \text{Ne}$

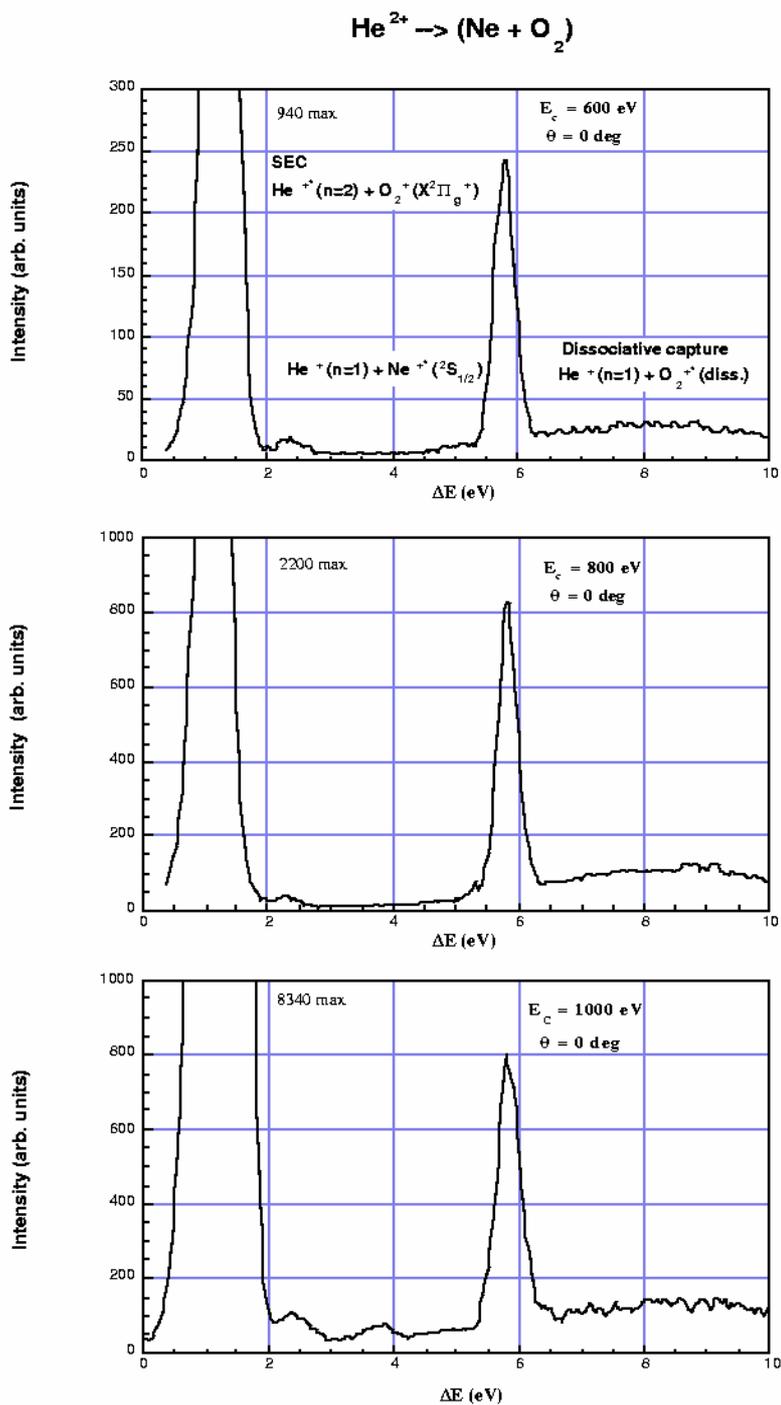
TES have been measured for impact energies of 600, 800 and 1000 eV at  $0^\circ$  scattering angle (see fig. 4). Apart from the calibration peak due to SEC from Ne, the dominant SEC channel involves an energy gain of 1.3 eV and can be identified as non-dissociative SEC



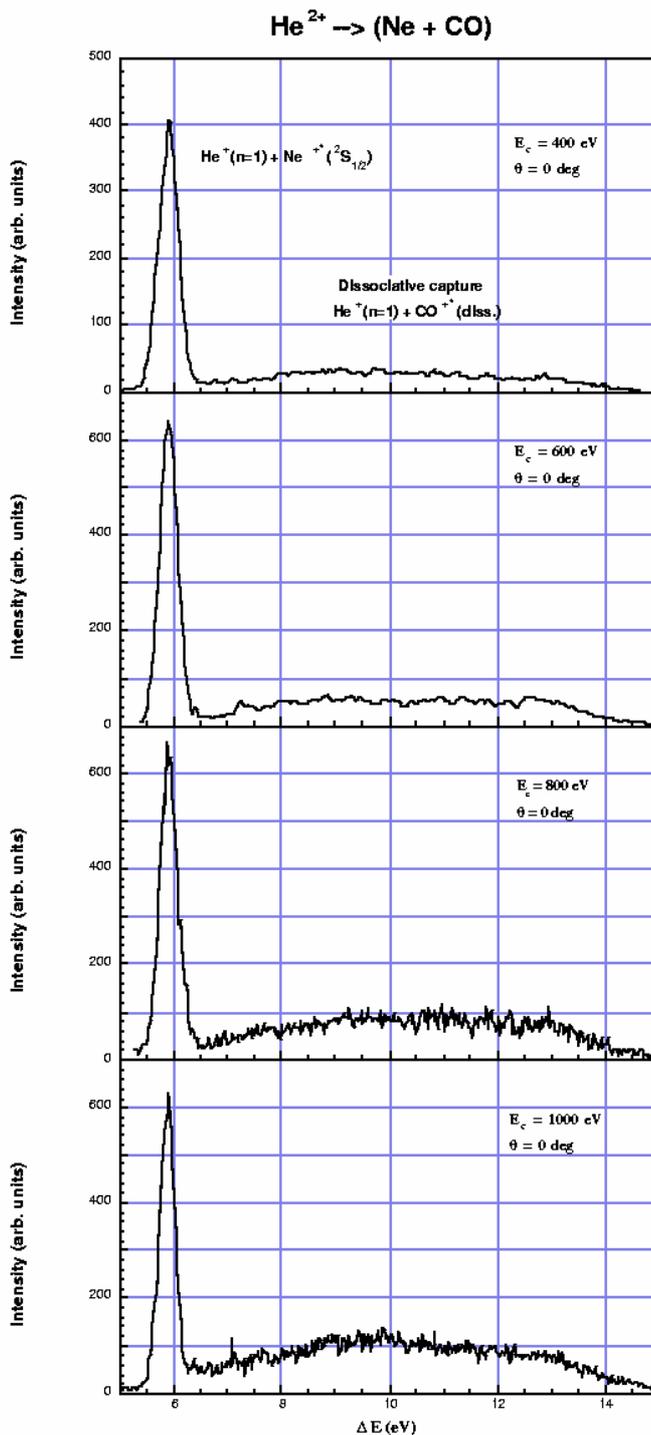
The rather broad feature for energy gains  $6 \leq \Delta E \leq 10$  eV corresponds to dissociative SEC into  $\text{He}^+(n=1)$ :



We have also measured TES for impact energies from 600 to 1000 eV at a scattering angle of  $0.2^\circ$ . The same reaction channels as mentioned above were observed, but for the finite scattering angle the non-dissociative SEC became slightly less important.



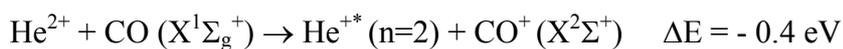
**Fig. 4:** TES for direct and dissociative SEC in He<sup>2+</sup> - O<sub>2</sub> collisions; the peak near 6 eV results from SEC in He<sup>2+</sup> - Ne collisions as utilized for calibration of the energy gain scale.



**Fig. 5:** TES for dissociative SEC in He<sup>2+</sup> - CO collisions; the peak near 6 eV results from SEC in He<sup>2+</sup>-Ne collisions as utilized for calibration of the energy gain scale.

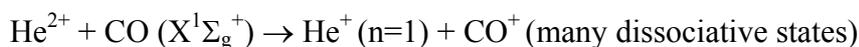
**He<sup>2+</sup> + CO / Ne**

For this collision system TES have been measured at impact energies between 400 and 1000 eV at 0° scattering angle (see fig. 5). Here the non-dissociative SEC channel

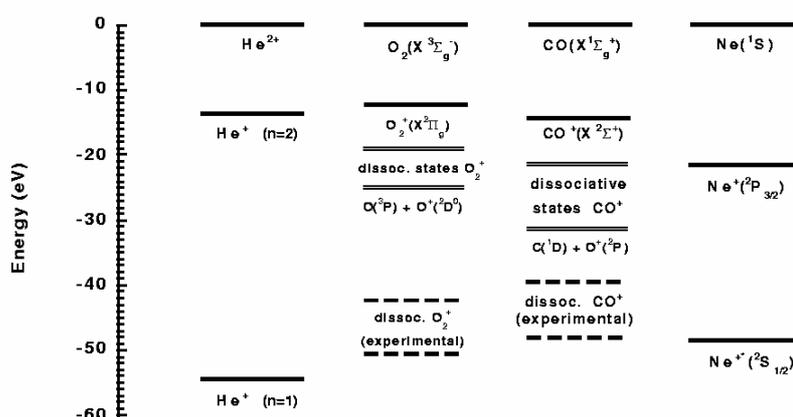


was practically not observable despite its small endothermicity, which is in clear contrast to similar measurements performed at only slightly higher impact energies [7].

Exothermic dissociative SEC channels



involve energy gains  $6 \leq \Delta E \leq 15$  eV. TES for scattering angles of  $0.2^\circ$  and  $1^\circ$  show similar importance of dissociative SEC with respect to SEC from Ne as for exact forward-scattering.



**Fig. 6:** Energy level diagram for SEC in collisions of  $\text{He}^{2+}$  with Ne,  $\text{O}_2$  and CO.

Fig. 6 shows energy levels of interest for both collision systems considered. On the left handside the relevant levels for SEC into  $\text{He}^{2+}$  are given, and on the right hand side levels are shown for SEC from Ne which was applied for calibration of the TES energy gain scale. In between we have indicated some energy levels for the target molecules  $\text{O}_2$  and CO and their singly charged ions. For  $\text{O}_2$  direct SEC into  $\text{He}^+(n=2)$  is exothermic whereas for CO it is endothermic. The endothermicity is even larger for direct SEC from  $\text{H}_2$  and  $\text{N}_2$  which explains why for these two collision systems no measurable TES peaks on the endothermic side could be found in our impact energy range. Furthermore, we have indicated known dissociative states for both molecular ions and the energy gain region where actually dissociative SEC has been observed ("experimental"). It is obvious that also for dissociative SEC, in this case into the  $\text{He}^+$  ground state, the slightly exothermic reaction channels are strongly preferred.

## Summary and outlook

By means of the translational attenuation method, a priori unknown metastable doubly-charged primary ion beam fractions can be characterized for subsequent single electron capture (SEC) reactions. The method utilizes the strong selectivity of SEC at low impact energies in suitably chosen attenuation gases, which causes preferential attenuation of either ground state- or metastable doubly charged ions before entering the translational energy spectrometer. The change in the primary ion

beam composition due to the selective attenuation can directly be observed in the resulting TES and serves for a clear interpretation of the latter. Measuring TES for different attenuation gas pressure permits the absolute determination of metastable ion beam fractions. Furthermore, TES measurements were made for non-dissociative and dissociative SEC in slow collisions of  $\text{He}^{2+}$  with  $\text{O}_2$  and  $\text{CO}$ . Our preliminary results highlight not only the relevance of the reaction window for "direct SEC" (no dissociation of the post-collisional molecular ions) from molecular target particles, but also show that a similar reaction window can be assumed for dissociative SEC. This reaction window is apparently shifted toward somewhat larger energy gains if compared with direct SEC. Further measurements along these lines and classical trajectory Monte-Carlo (CTMC-) calculations are in progress for a better understanding of the dissociative SEC.

### Acknowledgments

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