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International Journal of Mass Spectrometry 233 (2004) 239–244

International Journal of Mass Spectrometry

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State-selective and total single-electron capture for impact of slow He^{2+} ($E \leq 1$ keV) on H_2, O_2 and CO

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Received 27 October 2003; accepted 21 December 2003

Abstract

Projectile state-selective single-electron capture for impact of slow He^{2+} (impact energy $\leq$ 1 keV) on H_2, O_2 and CO has been studied experimentally by means of translational energy spectroscopy. Comparative measurements were made with a Ne gas target for absolute calibration of the electron capture cross-sections for the molecular targets. We compare the importance of "direct" single-electron capture (transition of one electron only without excitation or dissociation of the ionised target molecule) versus single-electron capture accompanied by target excitation (transfer excitation, possibly leading to molecular dissociation). Our measurements are extended to lower impact energies than earlier ones and are qualitatively interpreted on the basis of Landau–Zener transitions in the collisional quasimolecules. Similar as for direct single-electron capture, transfer excitation proceeds preferentially within a "reaction window" for the involved reaction energy defect.

Keywords: Single-electron capture; Slow He^{2+} ions; Electron capture cross-sections; Translational energy spectroscopy

1. Introduction

Impact of slow ($\leq$1 keV/amu) multicharged ions on neutral particles (atoms, molecules) influences the radiation balance of optically thin non-equilibrium plasmas as, e.g., the scrape-off layer, edge and divertor regions of tokamak fusion plasmas [1], or in astrophysical and ionospheric environments [2]. The electrons can be captured into excited projectile states which subsequently decay by photon emission [3]. However, in competition with single-electron capture involving transitions of only one electron (SC), transitions of more than one electron can take place to produce doubly excited projectiles (double-electron capture, DC) being subject to autoionisation (transfer ionisation, TI), or give rise to ionisation and excitation of the target particles (transfer excitation, TE) [4]. Classical, semiclassical and fully quantum–mechanical treatments of SC processes are well advanced [3,5], but the theoretical description of many-electron transitions as DC, TI and TE is not so far developed. Therefore, experimental studies of electron capture with many-electron transitions may help to improve their theoretical description. Electron capture from molecular targets is even more complex. In this context the He^{2+}–H_2 system may be regarded as a model case for both experimental and theoretical considerations (for a recent survey, cf., Ref. [6]). In addition, such collisions are of direct interest for fusion edge plasmas [1]. At impact energies E below a few 100 eV/amu, DC [7,8] and also TE [9] become more probable than SC, which is due to the small endothermicity of SC into the He^{+} ($n = 2$) state (−1.8 eV, see Fig. 1). In contrast, SC from O_2 is slightly exothermic (+1.5 eV, cf., Fig. 1) and therefore stays more important than TE down to considerably lower impact energy [10]. Finally, in He^{2+}–CO collisions SC is again slightly endothermic (−0.4 eV, cf., Fig. 1) and therefore we expect a similar behaviour of SC versus TE cross-sections as for He^{2+}–H_2 collisions, which was already demonstrated in previous work ([11] and Refs. therein).

We present a comparative study for these three collision systems at He^{2+} impact energies $E \leq 250$ eV/amu by means of translational energy spectroscopy (TES [12]). Emphasis is given to the importance of SC versus TE. Our experimental results extend to somewhat lower ion

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doi:10.1016/j.ijms.2003.12.036

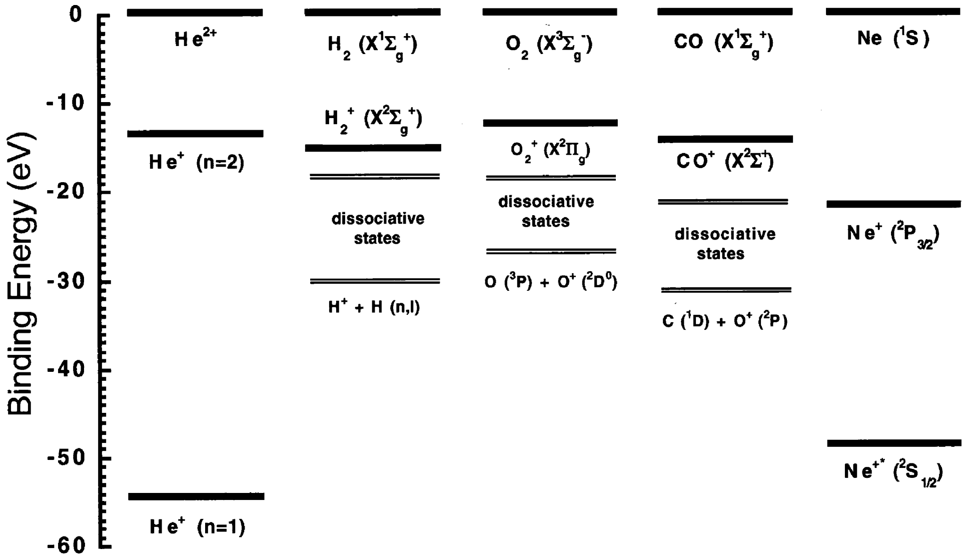

Fig. 1. Binding energy levels for single-electron capture by He^{2+} from H_2, O_2, CO and Ne. For molecular targets the regions indicated as "dissociative states" refer to tabulated dissociative molecular states, whereas in our measured TES (cf., Figs. 2–5) the observed TE features (cf., text) correspond to much higher excited dissociative states.

impact energies than for published data and are qualitatively interpreted with simple models for single-electron capture in collisions of slow multicharged ions with neutral particles.

2. Experimental methods and presentation of results

Translational energy spectroscopy determines the kinetic energy change of projectile ions which are (nearly) forward-scattered in collisions with neutral target particles of interest. In this specific scattering geometry the projectile assumes almost completely the inelastic energy loss or gain ΔE from the respective reaction channel [12], and different reaction channels can be identified from the resulting translational energy spectra. Slight deviation of the projectile scattering angle from the forward direction discriminates large impact parameter collisions and prefers reaction channels with transitions at smaller internuclear distance.

For our present experiments, He^{2+} primary ions were extracted from a 5 GHz ECR ion source [13], magnetically mass-to-charge analysed and guided into a translational energy spectrometer which has been described in detail elsewhere [12]. Electrostatic lenses for ion beam deceleration and focusing and a hemispherical monochromator define the projectile ion impact energy in front of the target gas beam. A second hemispherical energy analyser with channel electron multiplier detector collects the post-collision singly charged He^+ ions with different energy changes. A kinetic energy resolution of typically 500 meV proved sufficient for clear distinction of the different single-electron capture channels in the here measured TES. Molecular target gases were prepared with a calibrated 20% (vol.) admixture of Ne, for which at low impact energy only one SC channel was found to be relevant (see Fig. 1):

$$He^{2+} + Ne \rightarrow He^{+}(1s) + Ne^{+*}\ (2s2p^6)^2S \qquad (\Delta E = 5.94\,eV) \qquad (1)$$

By comparing relative TES peak heights for single-electron capture from Ne and the selected molecular gas targets (H_2, O_2, CO), we can directly evaluate the different state-selective electron capture cross-sections for the molecular gas targets with respect to the total cross-section for single-electron capture from Ne. This is possible because at low impact energy only the state-selective SC reaction (1) contributes to the total single-electron capture in the He^{2+}–Ne system (we note that total electron capture cross-sections can be measured in a simpler way than state-selective ones). Moreover, the well-defined projectile energy gain for SC from Ne serves for calibrating the TES energy change scales for single-electron capture from the molecular target species.

In addition to such TES measurements, total cross-sections for single-electron capture from the molecular target gases have been measured at higher impact energies ($E \geq 4\,keV$). This was done in a separate setup with a differentially pumped attenuation gas cell followed by electrical field deflection plates. The doubly charged, singly charged and neutral projectile fluxes behind the target gas cell were measured in shielded Faraday cups.

For detection of the neutral component we calibrated respective electron yields for He^+ ion impact with appropriate correction for additional potential electron emission [14]. The gas pressure in the target gas cell was measured with an absolutely calibrated capacitance manometer gauge.

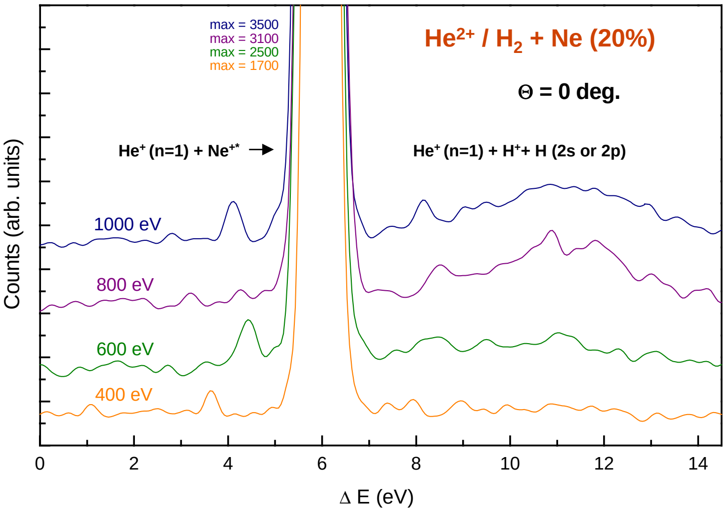

Fig. 2. Translational energy spectrum for single-electron capture in He^{2+}–H_2 collisions for scattering angle 0° and different impact energies. A 20% (vol.) admixture of Ne to the target gas gives rise to the large peak at $\Delta E = 5.94$ eV with relative size indicated on its left-hand side.

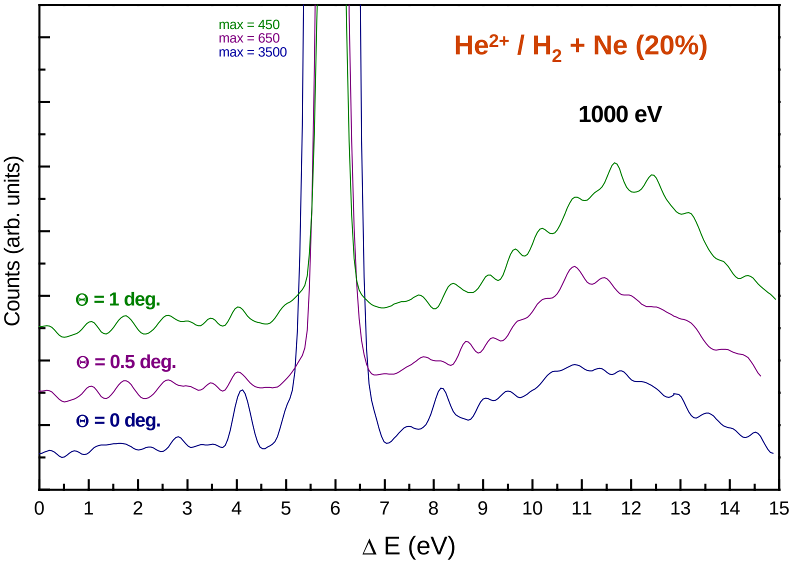

Fig. 3. Translational energy spectrum for single-electron capture in He^{2+}–H_2 collisions for different scattering angles and 1 keV impact energy. A 20% (vol.) admixture of Ne to the target gas gives rise to the large peak at $\Delta E = 5.94$ eV with relative size indicated on its left-hand side.

We performed these measurements with pure molecular target gases, pure Ne and molecular target gases with 20% (vol.) Ne admixture, respectively, keeping the target gas pressure sufficiently low for single collision conditions. Such total cross-section measurements were only made for comparison with respective published data, but we will extend them down to the same impact energies as for the here presented TES measurements. In particular, total single-electron capture cross-sections will be measured for He^{2+}–Ne collisions in order to calibrate similar data for the molecular target gases (see above). Typical TES are shown in Figs. 2 and 3 for H_2, Fig. 5 for CO and Fig. 6 for O_2, respectively. The figures exhibit raw (non-smoothed) experimental data. Comparisons with SC from Ne are shown in Figs. 4 and 7.

3. Discussion

3.1. He^{2+}–H_2

TES measured for $400 \leq E \leq 1000$ eV at 0° scattering angle in Fig. 2, and for $E = 1000$ eV and scattering angles of 0, 0.5 and 1° in Fig. 3 exhibit a rather small single-electron capture cross-section for H_2 in comparison with Ne (see also Fig. 4). We found no significant contribution on the endothermic side ($\Delta E < 0$) as observed at higher impact energy [9]. The broad unstructured feature on the exothermic side is related to TE via reactions

$$He^{2+} + H_2 \rightarrow He^{+}(1s) + H_2^{+*} \quad (\Delta E \geq 7\,\text{eV}) \tag{2}$$

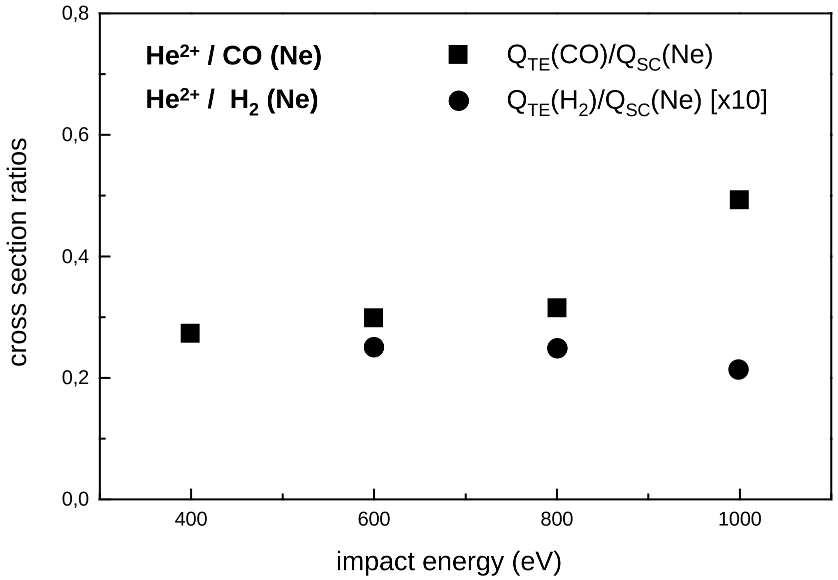

Fig. 4. Ratios of TE cross-sections Q_{TE} for single-electron capture from H_2 and CO, respectively, to the single-electron capture cross-section Q_{SC}(Ne) for Ne (cf., text). Note that the H_2 data has been enhanced by a factor of 10. The size of the symbols corresponds to statistical errors from reproducibility of measurements.

Quantum–chemical calculations (J. Harvey, University of Bristol, private communication) exhibit many dissociative $H_2{}^{+*}$ states corresponding to the here observed range of reaction energy defects. At our low impact energies SC reaction channels prefer considerably smaller exothermicity (typically $\Delta E = 5\,\text{eV}$ [12]) than the here observed maximum around 10 eV for the TE reactions (2), which corresponds to quasimolecular curve crossing radii $R_c \leq 3$ a_o [3]. Apparently, TE reactions (2) involve electronic transitions at considerably smaller internuclear distances than SC, corresponding to larger scattering angles. This explains discrepancies in total single-electron capture cross-sections measured by different groups (cf., discussion in Ref. [6]). As Fig. 3 shows, the TE feature becomes relatively more prominent with respect to SC from Ne if scattering deviates from the forward direction. Since at our impact energies the total cross-section for single-electron capture in $He^{2+} + H_2$ collisions is exclusively due to TE reactions (2), in our measurements the take-off angle for the charge-exchanged $He^+(1s)$ ions has been rather critical.

To elucidate this influence in somewhat more detail, we have modelled single-electron capture trajectories in colli-

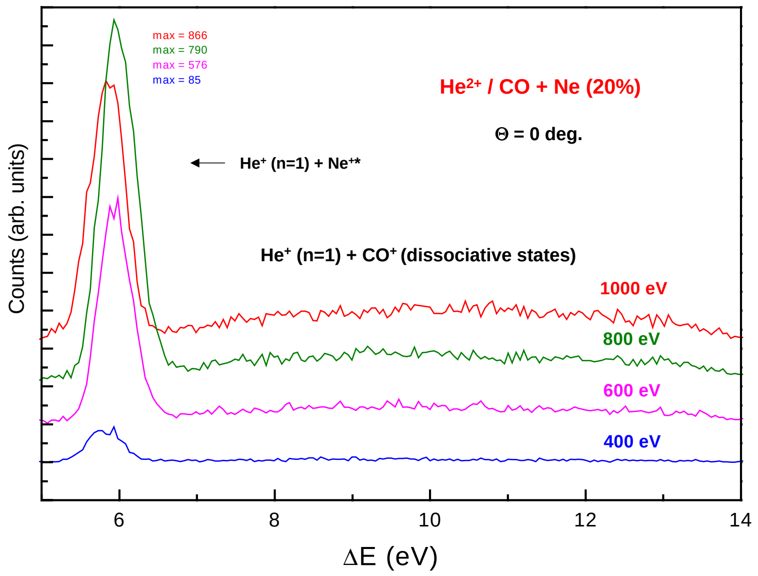

Fig. 5. Translational energy spectrum for single-electron capture in He^{2+}–CO collisions for scattering angle 0° and different impact energies. A 20% (vol.) admixture of Ne to the target gas gives rise to the large peak at $\Delta E = 5.94\,\text{eV}$ with relative size indicated on its right-hand side.

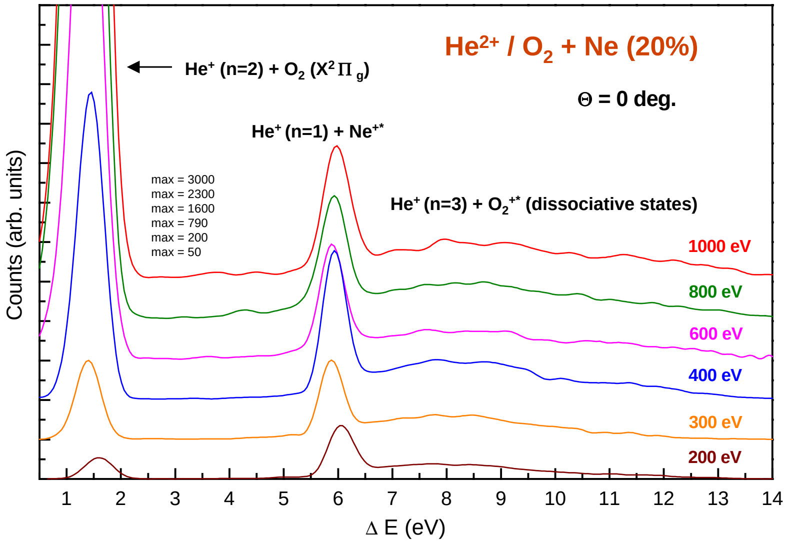

Fig. 6. Translational energy spectrum for single-electron capture in He^{2+}–O_2 collisions for scattering angle 0° and different impact energies. A 20% (vol.) admixture of Ne to the target gas gives rise to the peak at $\Delta E = 5.94\,\mathrm{eV}$. The dominant peak at $\Delta E = 1.5\,\mathrm{eV}$ with relative size indicated on its right-hand side is related to DC from O_2 (cf., text).

sions of He^{2+} with H_2. We took into account the polarisation attraction on the incoming trajectory until the "active electron" is captured at a certain crossing radius R_c. The result showed that for our TES acceptance angle of $\pm 0.5°$ at an impact energy $E = 1000\,\mathrm{eV}$ the $\mathrm{He}^{+}(1s)$ projectiles will not be detected if the electron is captured at $R_c = 2.5a_0$. Fig. 4 compares the Ne- and H_2-related TES features (areas under the respective "peaks" integrated), from which we learn that at $E = 1000\,\mathrm{eV}$ the total single-electron capture cross-section for Ne is about 40 times larger than for H_2.

3.2. *He^{2+}–CO*

Inspection of Fig. 5 shows that for single-electron capture by He^{2+} from CO the situation is similar as for H_2 (see above). We found no indication for SC into He^{+} ($n = 2$) states which would constitute the least endothermic SC channel (cf., Fig. 1), in contrast to the situation at higher impact energy [11]. The broad feature on the exothermic side can be related to TE reactions

$$\mathrm{He}^{2+} + \mathrm{CO} \rightarrow \mathrm{He}(1s) + \mathrm{CO}^{+*} \quad (\Delta E > 7\,\mathrm{eV}) \tag{3}$$

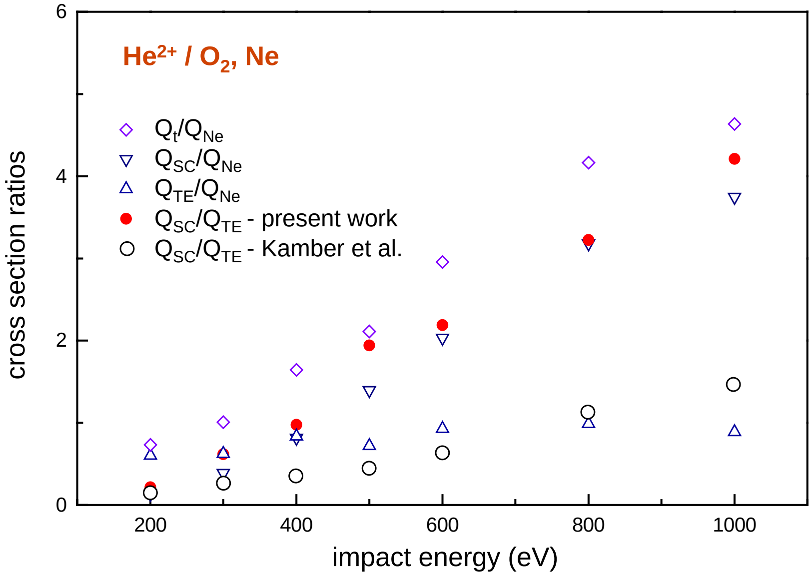

Fig. 7. Ratios of SC and TE cross-sections Q_{SC} and Q_{TE}, and of their sum Q_t for single-electron capture by He^{2+} from O_2, respectively, to the single-electron capture cross-section Q_{Ne} for Ne (cf., text). Furthermore, we compare the ratios $Q_{\mathrm{SC}}/Q_{\mathrm{TE}}$ from our TES measurements (see Fig. 6 with respective data from Ref. [10]). The size of the symbols corresponds to statistical errors from reproducibility of measurements.

Fig. 4 compares the Ne- and CO-related TES features exhibited in Fig. 5, which shows that at 1 keV He^{2+} impact energy the total single-electron capture cross-section for CO is only about a factor of two smaller than for Ne, still almost exclusively due to TE (see also Ref. [11]).

3.3. He^{2+}–O_2

As Fig. 6 shows, here the situation is markedly different from the cases for H_2 and CO, because SC can now proceed in the slightly exothermic channel (4a), see also Fig. 1. Consequently, we observe a dominant feature for this reaction

$$\mathrm{He}^{2+} + \mathrm{O}_2 \rightarrow \mathrm{He}^{+}\,(n = 2) + {\mathrm{O}_2}^{+} \quad (\Delta E = 1.5\,\mathrm{eV}) \tag{4a}$$

On the exothermic side of the peak related to SC from Ne, we find a broad feature which is ascribed to TE reaction channels as for the two other molecular target species

$$\mathrm{He}^{2+} + \mathrm{O}_2 \rightarrow \mathrm{He}^{+}(1\mathrm{s}) + {\mathrm{O}_2}^{+*} \quad (\Delta E > 7\,\mathrm{eV}) \tag{4b}$$

In Fig. 7, we compare the cross-sections for reaction channels (4a) and (4b) with the one for (1), as well as the respective total single-electron capture cross-sections (4a) + (4b) with (1). At 1 keV He^{2+} impact energy the total single-electron capture cross-section for O_2 appears to be about five times larger than for Ne. We can compare our results with recent data from Kamber et al. [10]. TES measurements published in [10] have been performed with a larger acceptance angle of ±1°. This explains the considerable discrepancy between the ratios of SC and TE contributions from both sets of data and clearly shows that our present TES measurements are affected by a too small acceptance angle. In order to overcome this considerable experimental difficulty, new TES measurements are planned with the projectile scattering angle varied in small steps around the forward scattering direction within ±2°. This will reveal how the respective relative SC and TE contributions depend on the scattering angle, and decide which take-off angle for the respective total cross-section measurements need to be assured at low impact energy. In the particular case of He^{2+}–H_2 collisions, double-electron capture (DC) total cross-sections become more important at low impact energy than SC and TE total cross-sections (cf., discussion in Section 1). For a precise determination of these total DC cross-section at low impact energy, we will conduct careful ion flux attenuation measurements after having obtained the respective SC and TE total cross-sections.

4. Summary

We have investigated “direct” electron capture (SC transition of one electron only) and single-electron capture involving transitions of more than one electron (TE) for impact of He^{2+} on H_2, CO and O_2 at impact energies $E \leq 1$ keV. For H_2 and CO only TE was found to contribute to single-electron capture, whereas for O_2 both SC and TE do contribute. As well known for SC reactions, apparently also TE reactions proceed preferentially within a “reaction window” for the related reaction energy defects, which however is shifted toward larger exothermicity than for SC reactions. Our relative cross-section measurements for SC and TE from molecular target gases have been linked to the SC cross-section for Ne which in our impact energy range is practically equal to its total single-electron capture cross-section. After the latter has been measured, it will give direct access to the absolute SC and TE cross-sections for the here studied molecular target gases.

Finally, comparison with data from other groups demonstrates a severe discrimination for TE cross-sections by our limited TES acceptance angle. For this reason, further TES measurements for different acceptance and (small) scattering angles will have to be carried out in order to produce reliable experimental data for comparison with theory.

Acknowledgements

This work has been carried out within Association EURATOM-OEAW. Its content is the sole responsibility of the authors and does not necessarily represent the view of the EU Commission or its Services. M.A. has been supported by the EU Network HPRN-CT-2000-00027 (MCInet).

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