Low-Energy State-Selective Charge Transfer by Multiply Charged Ions

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We present a combined rf-guided ion beam and photon emission spectroscopy method, which facilitates state-selective charge-transfer measurements at energies of direct relevance for astrophysics and fusion-plasma diagnostics and modeling. Ion energies have been varied from 1000 eV/amu down to energies as low as 5 eV/amu. Absolute state-selective cross sections have been obtained for He^{2+} and N^{5+} ions colliding on molecular hydrogen. Orders of magnitude differences are found between theory and the present results. This indicates clearly that such data are valuable as benchmarks for the necessary advancement of theoretical descriptions.

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Charge transfer dynamics at low energies is not only interesting from a fundamental atomic physics point of view, but, in practice, it is highly relevant for the charge state balance and light emission of plasmas. A prominent photon source is electron capture by multiply charged ions A^{q+} from neutrals B, because mainly excited states $A^{(q-1)+}(nl)$ are populated (the electron is captured nearly resonantly), which subsequently decay through photon emission. This sequence of processes is given by

$$A^{q+} + B \rightarrow A^{(q-1)+}(nl) + B^{+}$$

and

$$A^{(q-1)+}(nl) \rightarrow A^{(q-1)+}(n'l') + h\nu\,.$$

From this it is clear that the photon emission spectrum can be regarded as a "fingerprint" of the nl-state-selective electron capture processes. Often temperatures in astrophysical and fusion (divertor) plasmas are in such a range that the interactions proceed with kinetic energies in the range of 0.1 to 100 eV/amu. In order to use the information contained in the intensity and spectral distribution of the light it is necessary to quantitatively understand the underlying processes [1]. For example, because of its profound impact on the Jovian magnetosphere, the plasma torus of Jupiter at the orbit of its moon Io has been studied extensively by both earth-based visible light spectroscopy and rocket-based (Voyager) vacuum ultraviolet spectroscopy. However, attempts to determine basic Io torus properties from these observations have not yet succeeded due to the deficiencies in the modeling. A major drawback is thought to be the inaccuracy of theoretical cross sections for state selective electron capture by multicharged ions (charge states 2 to 5 [2]). A similar situation is encountered in the description of vacuum ultraviolet (VUV) emission from the tails of earth-passing comets such as Hale-Bopp and Hyakutake [3–5]. The emission results from electron transfer between multicharged solar wind ions (O, He, and Ne) and neutrals in the cometary tails. Accurate knowledge of the underlying processes would not just allow for explaining the observations but might enable the use of the VUV emission as a diagnostics tool for charge state and flux distributions of the solar wind ions. High resolution soft-x-ray and VUV spectra are expected to become routine with the Chandra and XMM (X-ray Multi-Mirror) observatories in operation.

When neutral particles are approached by multiply charged ions they become strongly polarized, leading to a mutual attraction which increases with the charge state of the ion. At collision energies above some 10 eV/amu this hardly influences the interaction dynamics. At lower energies it has dramatic consequences. It means that the collision trajectories are strongly influenced and that large impact parameters can result in relatively small distances of closest approach. This implies that cross sections for electron transfer strongly increase with decreasing collision energies. In a classical approach the electron capture cross sections increase with the charge state of the ions and inversely proportional to the velocity. Quantum mechanically even more interesting features are predicted to occur which are connected with the so-called orbiting resonances: At specific collision energies charge transfer proceeds via discrete rovibrational states of the transient molecular ion [6].

In general the numerous theoretical predictions of state-selective charge transfer cross sections well below 1000 eV/amu could be tested only partly or not at all. Furthermore, different calculations are frequently not in accordance with each other. A typical example is electron capture by O^{3+} (e.g., [7]) deriving its interest from the use of OIII emission to determine properties such as temperature and density of planetary nebulae and HII regions.

We present an experimental method, which facilitates state-selective charge-transfer measurements at energies of direct relevance for astrophysics and fusion-plasma diagnostics and modeling. The method combines the technique of rf ion guiding with crossed-beam photon emission spectroscopy. A schematic layout of the setup is depicted in Fig. 1. Details of the experiment and experimental procedures are presented elsewhere [8]. Briefly, ions extracted at 3.5 kV from the electron cyclotron resonance (ECR) ion source installed at the KVI atomic physics facility are injected via a five-element lens system into a rf

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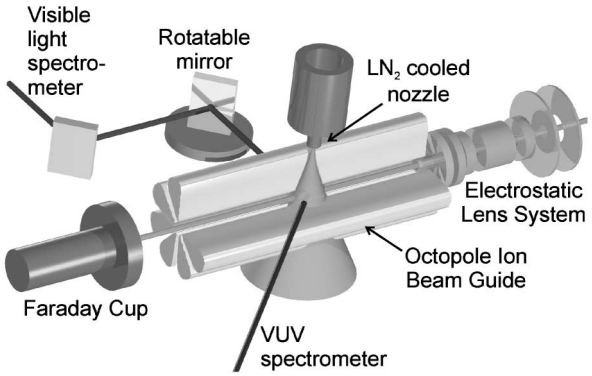

FIG. 1. Schematic representation of the apparatus of which only six of the eight rf guiding poles are shown.

multipole ion guide [9], in our case an eight-pole system. The method of rf multipole ion guiding was pioneered by Teloy and Gerlich [10] for singly charged ions and for the first time applied to multicharged ion beams by Okuno *et al.* [11]. Our rf octopole is a far more open system to allow for injecting a neutral target beam and observing the photon emission resulting from charge-transfer reactions. Because of possible field penetration, the openness of the system prevented the standard usage of rods as rf poles and special shaped, rigid 10-cm-long poles were designed and made by spark erosion.

The energy at which the ions interact with the target is defined by the difference between the source potential and the dc potential at which the octopole is floated (plus an offset due to the plasma potential of the source, ~15 V [8]). Down to a beam energy of basically 0, the ion beam can be guided through the system without intensity loss. However, there is a lower limit for performing well-defined experiments which is set by the energy spread of the primary beam. For our ECR ion source typical spreads are of the order of $5 \times q$ eV [8]. For N^{5+} ions this translates to an energy spread ~1.6 eV/amu. This sets the lower limit to ~5 eV/amu. In combination with a source from which ions are extracted with smaller energy spreads experiments can be performed at lower energies. As such a source, a recoil ion source might be considered which can produce cold ions (~0.1 eV/amu [12]).

The photon emission following charge-transfer reactions can be observed by two monochromators on opposite sides of the interaction region. The positioning is similar to the one used previously in our lab (e.g., [13]). The VUV spectrometer, which is used in the present experiments, covers a spectral range from 5–80 nm and is mounted under the double-magic angle to cancel polarization effects. The VUV system is calibrated absolutely on wavelength and sensitivity by cross-reference measurements on systems with well-known cross sections [13]. The spectrometer is equipped with a position-sensitive detector detecting a spectral range of almost 20 nm in one measurement.

To show the potential of the method we discuss results which were recently obtained for the He^{2+}-H_2 and N^{5+}-H_2 collision systems. Inelastic collisions of He^{2+} have attracted a lot of attention in connection with the modeling of He ash removal in the divertor region of tokamaks [14]. A particular point of interest is the charge state of the helium. If it is readily neutralized it is no longer magnetically confined and it may diffuse back into the core plasma thereby deteriorating the plasma. Total charge changing cross section measurements [15] showed that while being of the same order of magnitude around 0.5 keV/amu, two-electron capture dominates over the one-electron transfer processes by a factor of 50 around 10 eV/amu. Dominance of two-electron capture over one-electron capture is very rare (to our knowledge the only other system is C^{4+}-He). Although underestimating the total charge changing cross sections by a factor of 2, molecular orbital (MO) calculations in the energy range of 10–1000 eV/amu (Shimakura *et al.* [16]) gave the correct ratios between single- and double-electron transfer. In addition the MO calculations predicted that at energies below 100 eV/amu where two-electron capture dominates, single excited He($1s3l$) states are populates. The strong population of these He($1s3l$) states has been invoked to explain the anomalously intense HeI($1s3l \rightarrow 1s2l'$) emission observed when helium plasmas are in contact with cold hydrogen molecules (e.g., [17,18]).

Figure 2 shows some representative spectra for He^{2+}-H_2 collisions at energies in the range of 4000 down to 8 eV/amu. Two strong peaks are observed at 60.8 and 58.4 nm. The peaks result, respectively, from the second order of the HeII($2p \rightarrow 1s$) transition at 30.4 nm (60.8 nm, labeled as II-a) and the HeI($1s2p \rightarrow 1s^2$) transition (58.4 nm, labeled as I-a). Note that transition II-a originates from one-electron capture while I-a is due to two-electron capture. It is seen that dominance of either of the peaks swaps from peak II-a at high energies to peak I-a at low energies. This seems to confirm that at energies below ~100 eV/amu two-electron capture dominates over one-electron capture. The HeI($1s2p \rightarrow 1s^2$) line emission cross sections follow the same energy dependence as the total two-electron capture cross sections of Okuno *et al.* [15] and Shimakura *et al.* [16] but are smaller by a factor of 10 and 5, respectively. In line with theory this implies that capture into He($1s2p$) is just a weak two-electron capture channel. However, in contrast to the MO calculations the He($1s3l$) states are barely populated: there is no appreciable intensity at the position of the HeI($1s3p \rightarrow 1s^2$) transition (53.9 nm); see Fig. 2. It is off note that the HeI($1s3p \rightarrow 1s^2$) is by far the strongest decay channel of the HeI($1s3p$) state; i.e., it has a branching ratio of 97% [19]. In addition capture into the He($1s3s$) and He($1s3d$) states is included in the HeI($1s2p \rightarrow 1s^2$) emission because they decay to the He($1s2p$) state.

From this it is concluded that two-electron capture proceeds mainly into the He($1s^2$) ground state or the He($1s2s\,^1S$) metastable state. In our opinion the latter

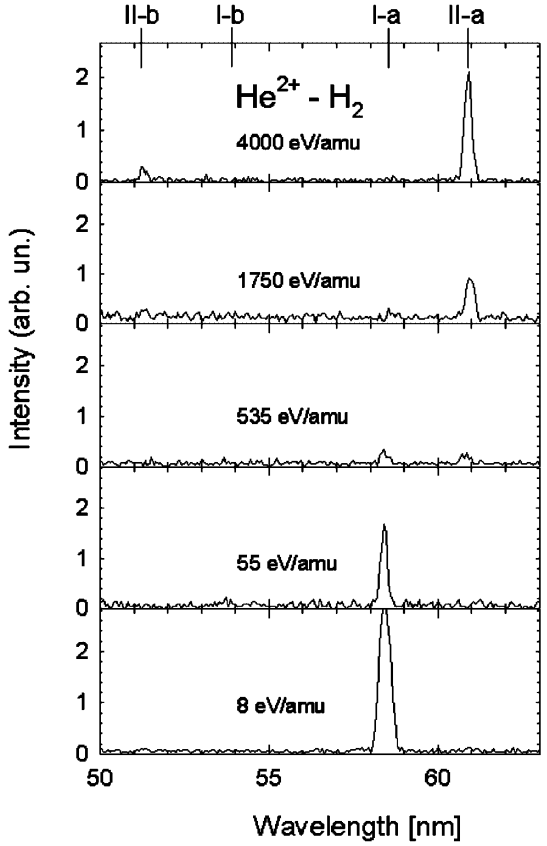

FIG. 2. VUV spectra from He^{2+}-H_2 collisions at energies ranging from 4000 eV/amu down to 8 eV/amu. Indicated are the following peak positions: II-a: second order of the HeII($2p \rightarrow 1s$) transition at 30.4 nm; II-b: second order of the HeII($3p \rightarrow 1s$) transition at 25.6 nm; I-a: HeI($1s2p \rightarrow 1s^2$) transition at 58.4 nm; I-b: HeI($1s3p \rightarrow 1s^2$) transition at 53.9 nm.

state seems the most likely, since the total amount of energy needed to remove the electrons from H_2 molecules is 50.8 eV (assuming Franck–Condon-type transitions). This should be compared with the binding energies of the He($1s^2$) and He($1s2s\,^1S$) states which are 79 and 58.4 eV, respectively. In addition from the potential-energy curves of the He^{2+}-H_2 system it is seen that the He($1s^2$) state can be populated only at small impact parameters [16]; therefore a large cross section is unlikely.

Next to hydrogenlike ions He-like ions are the most commonly used highly charged ions in theoretical collision studies (e.g., [20]). To show the value of the method to produce benchmark data for such collision systems, results for N^{5+}-H_2 are presented in Fig. 3. The cross sections for the $N^{4+}(3l)$ states, which are the dominantly populated states [21] are determined from the following emission lines: ($3p \rightarrow 2s$) at 20.9 nm, ($3d \rightarrow 2p$) at 24.8 nm, and ($3s \rightarrow 2p$) at 26.6 nm. All three transitions have branching ratios of 100%. It can be seen that at the higher energies ($\sim$1 keV/amu) there is good agreement with theory, but for lower energies theory and experiment start to diverge, leading to unexpectedly large discrepancies. For example, around 10 eV/amu the measured $N^{4+}(3s)$ cross sections exceed the theoretical ones by almost 4 orders of magnitude. Instead of a virtually closed channel, the $N^{4+}(3s)$ state turns out to be the main charge transfer channel. For the $N^{4+}(3p)$ and $N^{4+}(3d)$ states the deviations are smaller. However, they are much larger than one would expect, for in the 0.5–20 keV/amu energy range one finds good agreement between experiments and all kinds of theories (e.g., [20]).

To get at least an intuitive idea about the possible reasons for the discrepancies we will discuss the primary

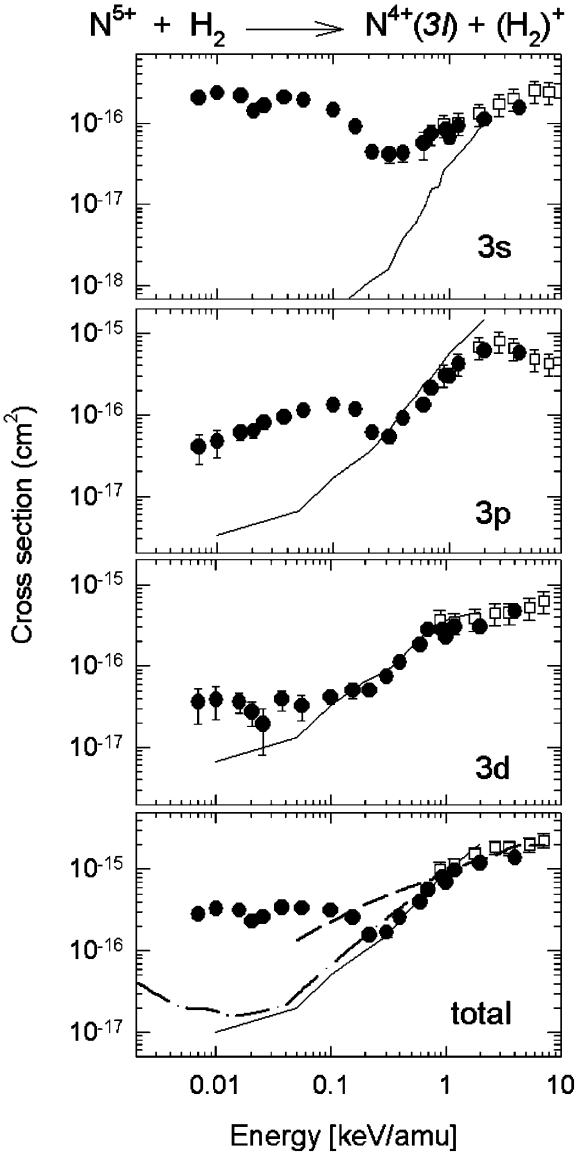

FIG. 3. Comparison of the present absolute state selective and total cross sections for N^{5+}-H_2 collisions (●) and other results (○: Dijkkamp *et al.* [21]; —: Kumar and Saha [22]; – · –: Gargaud and McCarroll [23]; and – –: Elizaga *et al.* [24]). The error bars (where exceeding the size of the dots) represent the statistical uncertainty. The systematic uncertainty of the measurements $\sim$20%.

collision process in terms of the extended over-the-barrier model [25]. In the framework of this model it is assumed that on the ingoing way of the collision the H_2 electrons subsequently transit the potential-energy barrier between the molecule and the N^{5+} ion. It is calculated that the least- and most-bound electron can transit the barrier at internuclear distances of 9.3 and 6.6 a.u., respectively. After the transitions the electrons occupy transient $(NH_2)^{5+}$ molecular states within the joint potential-energy well of the collision partners. For impact parameters $6.6 \leq b \leq 9.3$ only one electron becomes molecular, while for $b < 6.6$ both electrons are active. The corresponding geometrical cross sections are, by coincidence, of similar magnitude, i.e., 4×10^{-15} cm^2. On the way out of the collision the electrons, which became molecular, are redistributed over the projectile and target. The electrons will populate states around the resonant transition energies. For one-electron processes ($6.6 \leq b \leq 9.3$) the resonant energy lies in the gap between the $N^{4+}(n = 4)$ and $N^{4+}(n = 3)$ levels. Since the population distributions, the so-called reaction windows [25], become narrower with decreasing collision energy it is to be expected that the cross sections get smaller at lower energies as seen from Fig. 3. For the two-electron processes ($b < 6.6$) it is found that the strongest bound electron can near-resonantly populate the $N^{4+}(3s)$ state while recapture of the loosely bound electron by the hydrogen target is resonant with the ground state of the H_2^+ molecule. In this way two-electron processes lead to one-electron capture. Only a small fraction (5%–10%) of the geometrical cross section for two-electron processes suffices to explain the measured one-electron capture cross sections. Therefore, we propose that the differences between theory and experiment (cf. Fig. 3) are due to two-electron processes, implying that models should include these processes in the calculation of one-electron capture cross sections.

In conclusion we developed a combined rf-guided ion beam and photon emission spectroscopy method to measure state-selective charge-transfer cross sections at energies of direct relevance for astrophysics and fusion-plasma diagnostics and modeling. As examples the systems of He^{2+} and N^{5+} ions colliding on H_2 have been discussed. The ion energy has been varied down to energies as low as 5 eV/amu. In combination with a recoil ion source, the energies might be lowered much further to approximately 0.1 eV/amu. Unexpected large differences (orders of magnitude) with theory have been observed implying that the present and future measurements are valuable as benchmarks for improving theoretical descriptions.

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