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Experimental and theoretical investigation of electron capture and target excitation in (1–20 keV)H^+–K collisions

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Abstract. A combined experimental and theoretical investigation of single-electron capture and K(4s←4p) resonance line emission in H^+-K(4s) collisions at impact energies of 1-20 keV is presented. Comparisons are made with other experimental results as well as semiclassical calculations. The results are also discussed in connection with similar work on H^+-Li(2s) and H^+-Na(3s) collisions.

1. Introduction

Inelastic collisions between H^+ and alkali atoms have been the subject of numerous extensive experimental and theoretical studies. Interest in these collision systems not only arises for technical reasons (e.g. production of metastable hydrogen beams via charge exchange, cf DuBois and Toburen (1985) or possible pumping mechanisms for VUV laser, Allan *et al* (1986)). Much theoretical effort has also been devoted to a satisfactory understanding of such processes (Kubach and Sidis 1981, Kimura *et al* 1982, Fritsch 1984). In view of the simplicity of such 'quasi-one-electron collision systems' they can serve well for a crucial test of advanced collision theories.

Though considerable progress has been achieved in the accurate description of the relevant processes, some uncertainties remained especially for electron capture from potassium by protons, as has been reported for example by Ebel and Salzborn (1987).

While total single-electron capture cross sections from different authors show similar energy dependences, their absolute values differ by almost 50%. On the one hand we find good agreement when comparing the experimental results of Nagata (1980) with the molecular orbital (MO) calculations of Kimura *et al* (1982), on the other hand atomic orbital (AO) calculations by Fritsch (1984) are in agreement with experimental data of Grüebler *et al* (1970), which however give larger total capture cross sections than those of Nagata (1980). Finally, the measurements of Ebel and Salzborn (1987) fall between both sets of experimental data and are close to the calculations of Kubach and Sidis (1981) which are of an atomic-orbital expansion type but do not include translation factors.

The differences in the experimental results might primarily be related to the target thickness determination for alkali vapours, as has been mentioned, e.g. by Berkowitz and Zorn (1984).

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(i) In vapour cell experiments usually the target thickness is determined by measuring the target cell temperature and deriving the corresponding equilibrium vapour pressure from standard literature sources. Apart from the relatively large uncertainties of vapour pressure data (typically 25%), additional errors may arise from determination of the effective target length.

(ii) In crossed beam experiments one can measure the effective alkali atom flux with a surface ionization detector if the mean velocity of the alkali atoms is known from vapour temperature measurements.

(iii) The absorption of resonant K I light in K vapour has been utilized for target density determination by Phelps *et al* (1979).

(iv) If reliable electron capture cross sections are known for a specific alkali target, an absolute target thickness determination can be circumvented by using these cross section data for calibration. Should there be future corrections to the reference data because of more accurate calibration measurements, experimental data obtained in this way can easily be rescaled. This last method has been used in all our previous investigations using Li and Na alkali targets (Aumayr and Winter 1985, Aumayr *et al* 1987a, b) and also in the measurements presented here.

Because of the above mentioned discrepancies in absolute cross sections for the total single-electron capture process

$$H^+ + K(4s) \rightarrow [H^0] + K^+ \tag{1}$$

further experimental as well as theoretical investigations were found to be of interest.

In the H^+ impact energy range considered (1–20 keV) excitation of the K(4s) valence electron primarily into the K(4p) state

$$H^+ + K(4s) \rightarrow H^+ + K(4p) \tag{2}$$

competes with electron capture into (excited) projectile states (mainly $H(n=2)$, cf Aumayr *et al* 1984b, Aumayr and Winter 1985). This competition might cause interference effects in the exit channels of the collision system, manifesting itself in an undulatory shape of cross sections against impact energy (cf Aumayr *et al* 1987a, b). Experimental data on reaction (2) are therefore also interesting for comparison with calculations.

2. Experimental results

Our experimental methods for determining absolute cross sections for reactions (1) and (2) have been presented in extensive studies of similar collision systems (cf Aumayr *et al* 1984a, 1987a, b, Aumayr and Winter 1985) and will only be described shortly.

Cross sections for total single electron capture in H^+–K collisions were obtained from the fraction of projectiles neutralized in a K vapour target cell under single-collision conditions. Calibration of the K target thickness was performed by rapidly changing between H^+ and He^+ projectiles, using the reference reaction

$$He^+ + K(4s) \rightarrow [He^0] + K^+ \tag{3}$$

for which cross sections have been accurately measured by McCullough *et al* (1978), using method (i) for target thickness determination (cf section 1).

The $K(4s \leftarrow 4p)$ emission cross section has been measured by means of photon spectroscopy of the K I $D_{1,2}$ 766.5 nm + 769.9 nm lines. Photons produced from crossed

H^+ ion and K(4s) atom beams were observed with an interference filter-photomultiplier system under the magic angle of 54.7° with respect to the ion beam axis, to avoid errors from possible anisotropic emission. Absolute emission cross sections have been determined by comparing the photon signals produced by electron and ion impact, respectively (cf Aumayr *et al* 1984a), utilizing the accurate electron impact emission cross sections of Phelps *et al* (1979) at 200 eV impact energy.

Our results are presented in figures 1 and 2 and listed in table 1. Total errors for single-electron capture cross sections as estimated to ±25% result mainly from the quoted error of the reference cross sections (±20%) and errors associated with H^0 flux measurements via particle induced electron emission (±10%, see discussion in Aumayr

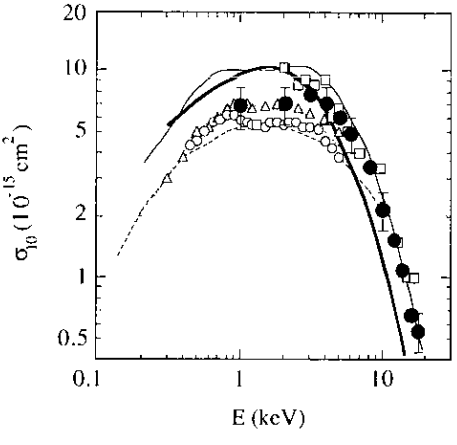

Figure 1. Total single-electron capture cross sections σ_{10} for impact of H^+ on K(4s) plotted against impact energy. Experimental results: ●, this work; □, Grüebler *et al* (1970); ○, Nagata (1980); △, Ebel and Salzborn (1987). Theoretical data are from: ——, this work; ——, Fritsch (1984); · · · ·, Kubach and Sidis (1981); – – – –, Kimura *et al* (1982).

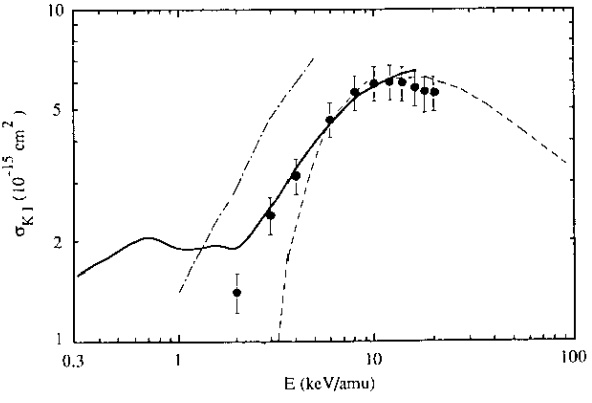

Figure 2. Experimental cross sections σ_{K1} for K(4s←4p) emission in inelastic H^+-K collisions plotted against impact energy: ●, this work. Theoretical data for K(4p) excitation by protons: ——, this work; · — · — ·, Kubach and Sidis (1981). Also shown are experimental cross sections for K(4s←4p) emission due to electron impact on K: – – – –, Phelps *et al* (1979). The electron energy is given in keV amu^{-1} to permit comparison with protons at equal impact velocity.

Table 1. Measured cross sections for total single-electron capture (σ_{10}) and K I($4s \leftarrow 4p$) emission ($\sigma_{K\,I}$) in H^+–K(4s) collisions.

E (keV)	σ_{10} (10^{-15} cm^2)	$\sigma_{K\,I}$ (10^{-15} cm^2)
1	6.77	—
2	6.96	1.41
3	7.68	2.40
4	6.88	3.17
5	5.87	—
6	4.96	4.68
8	3.40	5.63
10	2.11	6.00
12	1.52	6.05
14	1.08	5.99
16	0.65	5.77
18	0.55	5.54
20	—	5.58
Statistical error	±5%	±5%
Total error	±25%	±15%

and Winter 1985). The reproducibility over different sets of measurements was typically ±5%. The K I $D_{1,2}$ emission cross sections involve systematic errors from possibly slightly different collision geometries for (H^+–K) and (e^-–K) impact (less than ±10%), reproducibility (±5%) and the quoted error of electron impact emission cross sections (±12%).

3. Calculations

In the impact energy regime considered in this article, the semiclassical close-coupling method with atomic orbitals is expected to be quite appropriate, even more so since total capture is dominated by distant collisions (Kubach and Sidis 1981). In this work we follow the theoretical description of H^+–K(4s) collisions by Fritsch (1984) except for the following points.

In the work by Fritsch (1984) a pseudopotential has been taken for the construction of potassium states. Such a choice has the distinct advantage that occupied states need not be considered, the lowest state in such a potential is the $4l$ state for all angular momenta $l \leqslant 3$. Moreover, the pseudopotential by Bardsley (1974) is known to reproduce the energy levels of ground and excited potassium states to high accuracy. On the other hand, pseudopotentials have an l-dependent core term which is large at the origin of the nucleus and decays exponentially (in the case of the potential by Bardsley) with increasing separation. In a consistent implementation of a pseudopotential, the l-dependent term requires special attention, including projection techniques of travelling orbitals at the hydrogen centre to states of good angular momentum with respect to the potassium centre. In the work by Fritsch (1984) such projection procedures have been avoided by adopting, for two-centre matrix elements, the form of the $l = 0$ term for all l values in the qualitative argument that capture occurs at separations where the l-dependent term has essentially vanished. In quantitatively reassessing such a neglect of l-dependence it was now realized that this procedure is not satisfactory.

Indeed, fixing the form of the l-dependent term to one of the other terms given by Bardsley (1974) for $l=1,2$ leads to considerable variations of capture probabilities for any given capture channel. We did not attempt a consistent implementation of pseudopotentials into the available computer code; in an atomic orbital description of collisions matrix elements have to be calculated anew along every trajectory and hence complications introduced by projection procedures may well be computationally expensive.

In this work, therefore, a model potential is used for the representation of potassium states. Since, unlike in the case of sodium, we know of no dedicated potassium model potential, we took the potential of the form proposed by Garvey *et al* (1975) but with its η parameter changed to the value of 3.84 (from 3.52) by fitting the experimental potassium 4s energy. Representations of the potassium 4s, 4p and 4d states have been constructed by diagonalizing the potassium Hamiltonian in a space of eight s, six p, and four d hydrogenic states with optimized charge numbers, and higher pseudostates from such a construction have been kept in the basis up to positive energies of 6 au (occupied states are not included in the basis). The basis states at the potassium centre are listed in table 2. At the hydrogen centre, all nine H($n=2,3$) states are included in the basis.

From the close-coupling calculations with such a two-centre basis set, the sum of transfer cross sections into H($n=2$ and 3) states is displayed in figure 1, the excitation cross section for potassium 4p states in figure 2.

Table 2. Basis of hydrogenic states (n, l, Z) used in the close-coupling calculations of this work at the potassium centre, cf text. For each orbital angular momentum l, states with all magnetic quantum numbers m are included in the basis. The energies ε_{nl} designate the binding energies of the outermost electron in the lowest potassium states, as computed from diagonalizing the potassium Hamiltonian in the space of basis states, ε_{nl}^{exp} are the experimental energies from the compilation of Bashkin and Stoner (1978).

n	l	Z	ε_{nl}	ε_{nl}^{exp}	n	l	Z
1	0	24.93	—	—	1	0	17.56
2	0	15.45	—	—	1	0	4.32
2	1	23.06	—	—	1	0	1.45
3	0	9.91	—	—	1	0	0.40
3	1	10.21	—	—	2	1	12.56
3	2	7.34	—	—	2	1	3.19
4	0	5.00	−0.1588	−0.1596	2	1	0.78
4	1	3.42	−0.0981	−0.1003	3	2	1.18
4	2	2.84	−0.0637	−0.0614	3	2	0.71

4. Discussion

In figure 1 our measured and calculated total single-electron capture cross sections are compared with all available theoretical and experimental data. Excellent agreement is found between our experimental data and those of Ebel and Salzborn (1987) in the overlapping impact energy region. The improvement in accuracy as compared with

the data of Grüebler *et al* (1970) is obvious. Both sets of data seem to be rather consistent above 5 keV impact energy. Only fair agreement is found with the experimental results of Nagata (1980), which are by about 30% lower than our measurements.

In spite of a critical analysis of the calculations by Fritsch (1984) in section 3, the newly calculated total capture cross sections (for the transitions into the $n=2$ and 3 shells of H) turn out to be very close to those of the earlier work around the cross section maximum. At energies beyond 3 keV, however, the results calculated in this work are below corresponding results by Fritsch (1984). The deviations between the cross sections from the AO and MO calculations by Kimura *et al* (1982) presented here are unusually large, by about a factor of two. One would expect that the two model calculations would predict total transfer cross sections, for such distant collisions, of similar size. The results from the small-size AO-type calculations by Kubach and Sidis (1981) are closer to the result of the MO study, but given the limitations of the study by Kubach and Sidis, there is little to be learned from such a comparison.

The measured total capture cross sections from this work, as well as those by Ebel and Salzborn (1987) fall around the cross section maximum roughly in the middle between the results from the MO and AO expansion calculations, and actually happen to agree closely with the curve by Kubach and Sidis (1981). At higher energies, the AO curve underestimates the data by up to 40%. Much of the deviation at high energies could be understood by assuming a $1/n^3$ dependence of partial capture cross sections to final shells with principal quantum number n and extrapolating the calculated $n=3$ shell population accordingly to higher n shells. Around the cross section maximum, the 30% deviation is still within the assumed errors of the measurements and an estimated typical error of the calculations, of 15%, based on experience from many investigations. Still, the considerable deviation between results from the two large-scale close-coupling calculations is puzzling.

As probably the weakest point of the model calculations of this work we would identify the choice of potassium potential, naturally such a weakness would manifest itself in unreliable transition cross sections in close collisions at low energy. In distant collisions which dominate the transfer cross sections at energies around the cross section maximum, however, one would generally not expect uncertainties of calculated cross sections of the order of a factor of two. Unfortunately, in the absence of an improved model potential of potassium it is not possible to make more definite statements within the scope of this work.

In figure 2 our measured K(4s←4p) emission cross sections are compared with the results of our AO+ calculation for K(4p) excitation and calculations by Kubach and Sidis (1981). Comparison between experiment and theory suffers somewhat from the unknown population of higher excited K I states, which via cascading contribute to the K(4p) population. The experimental K(4s←4p) emission cross section of course includes such cascade contributions. However, Kubach and Sidis (1981) explicitly stated in their paper that their cross sections denoted as K(4p) do not refer to direct K(4p) excitation only, but merely represent '*all inelastic collision processes other than capture into H(n=2) states*'. Their data may therefore be considered as an upper limit to the K I emission cross section in fair agreement with the experimental results. Our calculations seem to agree well with our experimental data points except at the lowest energies where the theory description becomes increasingly sensitive to deficiencies of both the potassium potential and the basis set. At higher energies beyond 10 keV one may expect that an improved description of 4p excitation would require an improved representation of higher excited states of K and the continuum. This seems to be

foreshadowed in a slight overestimation of the experimental 4p excitation cross section by the calculations around 15 keV.

Cross sections for K(4s←4p) emission caused by electron impact (Phelps *et al* 1979) are also given in figure 2. Comparison is made for equal impact velocities of electrons and protons. Above 6 keV amu^{-1} ($v \approx 0.5$ au) the experimental proton impact excitation cross section joins smoothly into the electron impact excitation cross section, which demonstrates the probable dominance of direct (Coulomb) excitation at higher impact energy. Below $v \approx 0.5$ au, however, proton impact excitation channels become increasingly subject to interaction with single-electron capture channels. Because of these quasimolecular transitions, K I excitation by protons is possible even at impact velocities below the corresponding electron impact threshold (excitation energy for K(4s←4p) ≈ 1.61 eV, corresponding to $v \approx 0.35$ au or about 3 keV amu^{-1}). A similar behaviour could already be observed for excitation of Li and Na (Aumayr *et al* 1987b).

In figure 3 our data for total single electron capture in H^+–K collisions are plotted together with our previous results for H^+–Li (Aumayr and Winter 1985) and H^+–Na (Aumayr *et al* 1987a) collisions. Figure 4 shows a similar comparison for the resonance line emission cross sections following target excitation by protons (for Li(2s←2p) from Aumayr *et al* 1984a and for Na(3s←3p) from Aumayr *et al* 1987a). The curves in figures 3 and 4 represent smoothed curves through experimental data points. The main structures in the energy dependence significantly exceed the experimental error bars and can be attributed to interference effects (cf below).

From figures 3 and 4 the following systematics can be deduced.

The absolute size of the single-electron capture cross sections increases while the cross section maximum shifts to lower impact energies when changing the target from Li via Na to K. This is a direct consequence of the involved reaction energy defects for capture into the primarily populated H($n = 2$) states ($\Delta E = 0.94$ eV for K, 1.74 eV for Na, and 1.99 eV for Li, respectively). For the target excitation the magnitude of the excitation cross sections (excitation energy of K(4p): 1.61 eV, Na(3p): 2.1 eV and Li(2p): 1.85 eV) are also following the involved reaction energy defects (cf figure 4).

Deviations from this simple dependence are only found for K I emission at impact energies below 5 keV. This might be attributed to details of the competition between the weak excitation channel and the strong transfer channel for each system.

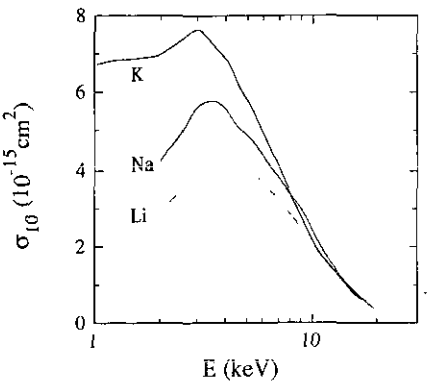

Figure 3. Comparison of measured total single-electron capture cross sections σ_{10} for impact of H^+ on Li(2s), Na(3s) and K(4s), respectively. Data for Li and Na from Aumayr and Winter (1985) and Aumayr *et al* (1987a), respectively.

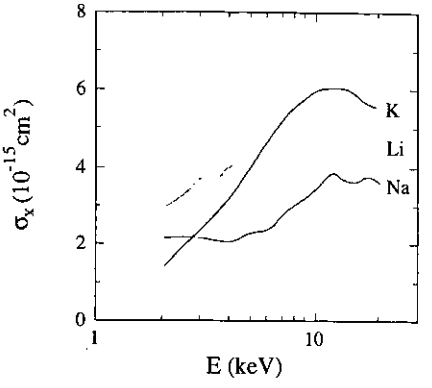

Figure 4. Comparison of measured absolute cross sections σ_x for target atom resonance line emission in collisions of H^+ with Li(2s), Na(3s) and K(4s), respectively. Data for Li(2s ← 2p) and Na(3s ← 3p) emission from Aumayr *et al* (1984a) and Aumayr *et al* (1987a), respectively.

In conclusion, a rather consistent picture for the total single-electron capture cross section in H^+-alkali collision systems is now present. Systematic alkali target species dependencies can be explained in a simple way with regard to reaction energy defects. For the H^+-K(4s) system, however, there are still unusually large and unexplained discrepancies between total capture cross sections from MO and AO expansion calculations around the cross section maximum.

Acknowledgments

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