

State-selective electron-capture measurements for N^{4+} -H and N^{4+} -H₂ collisions

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(Received 16 May 1997)

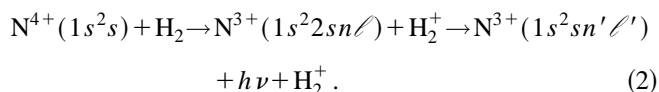
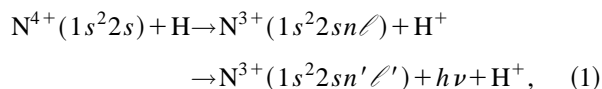
State-selective electron-capture cross-section measurements in the energy range between 1 and 4 keV/amu are reported for collisions between N^{4+} ions and atomic and molecular hydrogen. The cross sections are measured in a crossed-beam experiment by means of photon emission spectroscopy. The singlet and triplet states are resolved. It is found that the capture probabilities are not statistically distributed over both spin states and depend strongly on the primary energy. For collisions with atomic hydrogen the agreement with fully quantum-mechanical calculations is in general good. The sums of the experimental results for capture into all the singlet and triplet states are in excellent agreement with previous total one-electron capture measurements. Additionally for molecular hydrogen we performed multichannel Landau-Zener calculations that are found to be in fair agreement with the experimental data except for the highest angular momenta states. [S1050-2947(98)00701-X]

PACS number(s): 34.70.+e, 34.10.+x

I. INTRODUCTION

Single-electron capture is the dominant inelastic process that occurs in collisions between multiply charged ions and neutrals in the low- and intermediate-energy regimes. With a high probability electrons are transferred (quasi)resonantly into excited states of the ion. Therefore single-electron capture plays an important role in the energy and charge state balance of both astrophysical and thermonuclear fusion plasmas. The photon emission resulting from the decaying product ions contains important information on plasma parameters such as ion densities and temperatures. However, to determine these plasma quantities detailed knowledge about the charge transfer processes is required and therefore the study of single-electron capture receives much attention.

In this paper we report state-selective cross sections for single-electron capture in the following reactions:



The experiments are carried out in the energy range between 1 and 4 keV/amu by means of photon emission spectroscopy (PES), i.e., by detecting the photons $h\nu$ emitted in the $N^{3+}(n\ell) \rightarrow N^{3+}(n'\ell')$ transitions.

Due to the possibly important role of N^{4+} -H charge changing collisions in plasma diagnostics and modeling [1] several experimental and theoretical studies on charge transfer in N^{4+} -H collisions have been undertaken. Up to now $N^{4+} + H$ collisions were studied experimentally by Huq *et al.* [2] and Folkerts *et al.* [3] who used the merged-beam

technique to obtain accurate total charge transfer cross sections. Furthermore, translational energy spectroscopy (TES) experiments by McCullough *et al.* [4] provided the first information on state-selective cross sections. Their measurements, however, could not resolve the singlet and triplet states.

Theoretically the N^{4+} -H system was studied by Butler and Dalgarno [5] using a two-state Landau-Zener approximation. The N^{4+} -H system was analyzed by Feickert *et al.* [6] in the framework of a molecular orbital (MO) model at much lower energies than the ones used in our experiment. Quantum mechanical calculations by Stancil *et al.* [1], which extend previous calculations by Zygelman *et al.* [7] and calculations by Shimakura *et al.* [8], which have been extended in a recent paper by Folkerts *et al.* [3], cover several decades of the energy spectrum starting from a few tenths of eV up to several keV. In the following these two extensive sets of calculations will be referred to as Stancil *et al.* [1,9] and Shimakura *et al.* [8]. Their results are generally in good agreement with the available experimental results, however, our present results provide a much more stringent test of these models since both the singlet and triplet states are resolved. Generally it is assumed that in keV/amu collisions the distribution over different spin states is statistical [7]. The calculations by Stancil *et al.* and Shimakura *et al.* predict a ratio between triplet and singlet capture clearly deviating from statistical. With our high-resolution PES measurements it is possible to test this theoretical prediction.

II. EXPERIMENTAL METHOD

A. Photon emission spectroscopy setup

The state-selective electron-capture measurements were performed in a new crossed-beam setup installed at the atomic physics facility of the KVI Groningen. The cross sections are determined by means of PES, i.e., photons resulting from the decay of the excited product ions are detected by a vacuum ultraviolet monochromator sensitive in the wavelength region between 10 and 80 nm. This monochromator is calibrated absolutely on wavelength and sensi-

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tivity; see, e.g., Hoekstra *et al.* [10,11] and references therein. The setup is equally configured as the one described in Ref. [11], however, the present experiment is equipped with an octupole ion trap in which the primary ion beams, which are produced at keV energies by an ECR ion source, can be decelerated down to eV energies [12]. But in the present series of measurements the octupole guide was not yet used to decelerate ion beams to avoid any influence on the ion beam quality and to verify that absolute cross sections can be measured. These cross sections are not only taken to verify theoretical predictions but they should also serve as a reference for future measurements with the rf guide in operation.

B. Hydrogen target

The (atomic) hydrogen target is provided by a Slevin-type radio frequency source [13]. In this source a discharge of molecular hydrogen can be sustained by feeding about 30 W of rf power, at a frequency of 27 MHz, into a coaxial cavity. The atomic hydrogen effuses into the collision chamber through an optically blind Teflon capillary, which is cooled by a cryodrive to a temperature of 80 K to reduce the hydrogen recombination rate at the Teflon surface even further. The dissociation degree D is typically in the order of 75% and is defined by

$$D = \frac{n_H}{n_H + n_{H_2}}. \quad (3)$$

This parameter is checked before and after every measurement by observing the He II($2p \rightarrow 1s$) line emission resulting from collisions of He²⁺ with a pure H₂ target or with a mixed H-H₂ target. This is a sensitive parameter for the dissociation degree since the cross sections for electron capture from H and H₂ are significantly different [11,14].

The signal S^{off} resulting from collisions on a pure molecular target is given by

$$S^{\text{off}} = C n_{H_2} \sigma_{H_2}, \quad (4)$$

With n_{H_2} the H₂ density, σ_{H_2} the He II($2p \rightarrow 1s$) emission cross section, and C a calibration factor. When the rf is switched on both collisions on H and H₂ contribute to the signal S^{on} , which is given by

$$S^{\text{on}} = C \left[(1-k)n_{H_2}\sigma_{H_2} + \frac{2}{\sqrt{2}}kn_{H_2}\sigma_H \right]. \quad (5)$$

The density of the H particles equals $(2/\sqrt{2})kn_{H_2}$ when a fraction k of the H₂ particles is dissociated. Since the mass of the H particles is half the mass of the H₂ particles, the velocity of the H particles is $\sqrt{2}$ times the velocity of the H₂ particles. Now it follows that the fraction of dissociated molecular hydrogen k equals

$$k = \left(\frac{S^{\text{on}}}{S^{\text{off}}} - 1 \right) \left(\sqrt{2} \frac{\sigma_H}{\sigma_{H_2}} - 1 \right)^{-1}. \quad (6)$$

The dissociation degree is related to k by

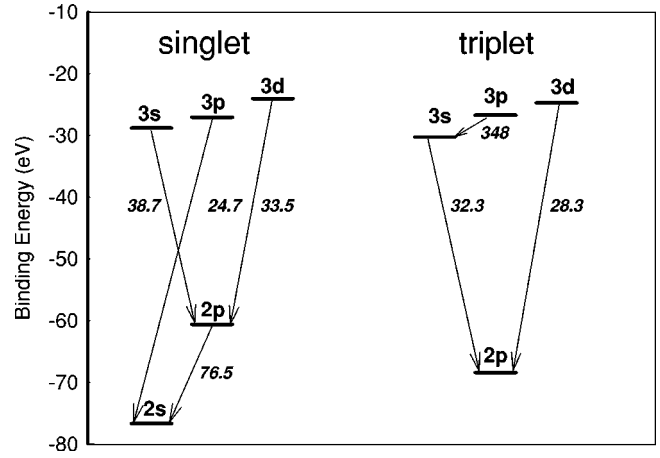


FIG. 1. N IV energy-level diagram for both the $3l$ singlet and triplet states. The wavelengths (in nm) of the transitions are indicated in italics.

$$D = \frac{\sqrt{2}k}{(\sqrt{2}-1)k+1}. \quad (7)$$

The H and H₂ densities are determined before and after each measurement of N⁴⁺ ions with a mixed H-H₂ target. Now the subtraction of the H₂ contribution to a spectrum resulting from collisions on a mixed target is straightforward. The H₂ target beam is calibrated absolutely against a static H₂ target. During the measurements the target pressure is kept low enough to ensure single collisions and is stabilized within 2%.

C. Photon detection

During collisions between N⁴⁺ ions and both atomic and molecular hydrogen the electrons are predominantly captured into the N³⁺($1s^2 2s 3\ell$) 1L and 3L states. Inspecting the relevant part of the Grotrian diagrams (see Fig. 1) it is clear that all relevant N IV($3\ell \rightarrow 2\ell$) transitions have wavelengths in the VUV, except for the ($3p \ ^3P \rightarrow 3s \ ^3S$) transition at 348 nm. Since this transition has a branching ratio of one, the observed emission of the ($3s \ ^3S \rightarrow 2p \ ^3P$) transition at (32.3 nm) results from capture into both the $3s \ ^3S$ state and the $3p \ ^3P$ state whereby the latter one contributes via the cascade ($3p \ ^3P \rightarrow 3s \ ^3S \rightarrow 2p \ ^3P$). The lifetimes for these states are so short [15] that all photons resulting from the decay of the reaction products are emitted in the viewing area of the VUV spectrometer. In principle also the other $3\ell \rightarrow 3\ell'$ transitions have to be taken into account. However, the branching ratios are so small [15] that they hardly contribute to cascade emission. This means that all emission cross sections can directly be converted into capture cross sections. The photon emission is observed with a VUV monochromator. This is a grazing incidence vacuum spectrometer positioned under the double magic angle, i.e., 54.7° with respect to the beam axis and tilted by 45°, to cancel polarization effects [11]. A position-sensitive microchannel plate detector enables simultaneous detection of lines within a range of 20 nm with a resolution of 0.6 nm. The wavelength-dependent sensitivity of the VUV monochromator has been determined absolutely with an accuracy of 20%

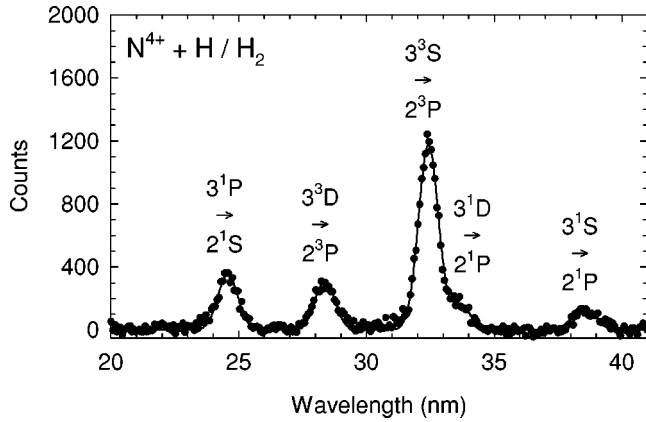


FIG. 2. Typical VUV spectrum resulting from N^{4+} ions colliding on a mixed H/H_2 target.

by means of various electron- and ion-impact processes with well-known cross sections. In the relevant spectral range (24 – 39 nm, see Fig. 1), the absolute calibration of the VUV system depends to a large extent on data for He^{2+} colliding on H_2 [14] and C^{4+} colliding on H_2 [10].

Figure 2 shows a typical PES spectrum of $N\text{ IV}$ emission in the VUV spectral range. The emission cross sections are related via

$$\sigma_{em}(nl \rightarrow mk) = \frac{4\pi}{\omega} \frac{q}{K(\lambda)Q} \frac{S(\lambda)}{nL} \quad (8)$$

to the measured signal $S(\lambda)$, where λ is the wavelength of the $nl \rightarrow mk$ transition, ω is the solid angle of observation, q the charge state of the ions, $K(\lambda)$ the quantum efficiency of the detection system, Q the accumulated charge, n the effective target density, and L the observation length.

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. One-electron-capture cross sections for N^{4+} colliding on H_2

The experimental results for electron capture into N^{4+} ions from collisions with molecular hydrogen are given in Table I and are depicted as a function of energy in Fig. 3.

TABLE I. Measured state-selective charge-transfer cross sections for N^{4+} ions colliding on H_2 molecules. The errors represent the relative errors only. The systematic absolute error is 20%.

| E (eV/amu) | $\sigma(3s\ ^1S)$ | $\sigma(3p\ ^1P)$ | $\sigma(3d\ ^1D)$ | $\sigma(3s\ ^3S) + \sigma(3p\ ^3P)$ | $\sigma(3d\ ^3D)$ |
|--------------|-------------------|-------------------|-------------------|-------------------------------------|-------------------|
| 860 | 5.85 ± 1.51 | | 3.04 ± 1.06 | | 2.05 ± 1.39 |
| 1140 | 4.53 ± 0.90 | 2.97 ± 0.52 | 2.01 ± 0.62 | 18.78 ± 2.85 | 1.81 ± 0.47 |
| 1290 | 4.19 ± 0.81 | 3.06 ± 0.56 | 2.34 ± 0.52 | 21.39 ± 3.67 | 3.07 ± 0.58 |
| 1470 | 3.32 ± 0.49 | 3.32 ± 0.49 | 1.89 ± 0.56 | 18.14 ± 2.21 | 1.89 ± 0.39 |
| 1570 | 5.30 ± 1.10 | 3.98 ± 0.75 | 2.74 ± 0.71 | 22.47 ± 3.90 | 3.16 ± 0.67 |
| 1710 | 3.95 ± 1.22 | 2.88 ± 0.81 | 1.85 ± 0.85 | 17.21 ± 4.27 | 2.39 ± 0.78 |
| 2000 | 3.07 ± 0.50 | 2.91 ± 0.36 | 1.62 ± 0.42 | 13.73 ± 1.48 | 2.49 ± 0.36 |
| 2290 | 3.03 ± 1.22 | 3.43 ± 1.05 | 2.04 ± 0.92 | 15.84 ± 4.30 | 3.11 ± 0.95 |
| 2570 | 3.35 ± 0.94 | 3.92 ± 0.84 | 3.14 ± 0.89 | 16.68 ± 3.36 | 3.77 ± 0.86 |
| 2860 | 3.32 ± 0.61 | 3.51 ± 0.44 | 2.14 ± 0.48 | 15.66 ± 1.71 | 4.09 ± 0.53 |
| 3430 | 2.52 ± 0.83 | 2.90 ± 0.69 | 1.70 ± 0.65 | 12.18 ± 2.63 | 3.81 ± 0.91 |
| 3710 | 2.35 ± 0.63 | 3.37 ± 0.63 | 2.22 ± 0.57 | 14.17 ± 2.48 | 4.46 ± 0.83 |

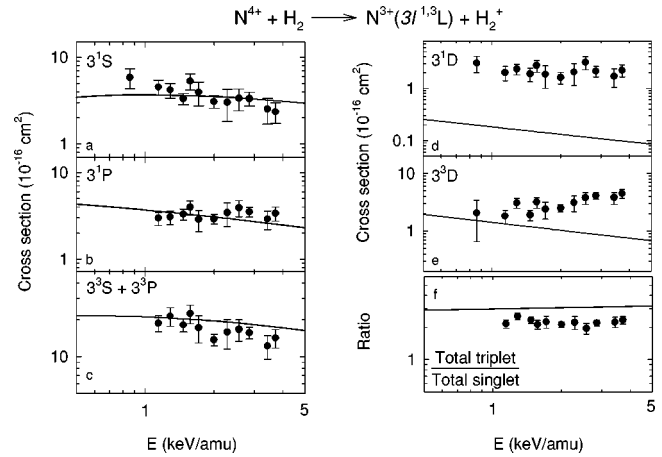


FIG. 3. Measured and calculated (MCLZ) state-selective charge-transfer cross sections for N^{4+} ions colliding on H_2 molecules.

The relative errors represent the statistical errors at a 90% confidence level and the uncertainties in the target density. The latter ones are due to possible fluctuations in the target density and the overlap of the ion and target beams. Apart from the relative errors a systematic error of 20% has to be included due to uncertainties in the absolute calibration of the VUV spectrometer.

Until now this collision system was hardly studied, i.e., only two experiments were performed. Wang and Church [16] measured a rate coefficient of 4.2×10^{-10} cm^3/s for electron capture at a mean energy of 2.3 eV and McCullough *et al.* [4] measured the translational energy gain of 24-keV N^{4+} ions colliding on H_2 molecules. Their spectrum shows capture into different ℓ states, but no capture probabilities were determined.

Since no theoretical calculations are available the multi-channel Landau-Zener (MCLZ) model is used to describe the general trends in the capture processes. In the model the electronic transitions occur at avoided crossings between adiabatic potential-energy curves and are induced by radial couplings only. The probability for a transition between an initial potential curve of state i to the potential curve of state j is given by $(1-p)$ with p given by

TABLE II. Measured state-selective charge-transfer cross sections for N^{4+} ions colliding on H. The errors represent the relative errors only. The systematic absolute error is 20–25 %.

| E (eV/amu) | σ ($3s \ ^1S$) | σ ($3p \ ^1P$) | σ ($3d \ ^1D$) | σ ($3s \ ^3S$) + σ ($3p \ ^3P$) | σ ($3d \ ^3D$) |
|--------------|-------------------------|-------------------------|-------------------------|---|-------------------------|
| 1140 | 4.73 ± 0.67 | 2.92 ± 0.94 | 2.74 ± 0.92 | 10.87 ± 3.51 | 3.89 ± 1.19 |
| 1470 | 1.18 ± 0.57 | 3.53 ± 0.51 | 2.96 ± 0.59 | 13.34 ± 1.93 | 2.92 ± 0.46 |
| 1570 | 2.35 ± 0.61 | 4.23 ± 0.78 | 3.49 ± 0.69 | 16.86 ± 3.17 | 4.76 ± 0.87 |
| 1710 | 2.07 ± 0.92 | 3.96 ± 1.15 | 2.94 ± 1.02 | 14.61 ± 4.18 | 4.78 ± 1.40 |
| 2000 | 1.33 ± 0.52 | 2.56 ± 0.57 | 2.67 ± 0.64 | 9.31 ± 2.04 | 4.24 ± 0.89 |
| 2290 | 1.95 ± 0.90 | 2.62 ± 0.80 | 3.21 ± 1.07 | 13.45 ± 3.80 | 4.96 ± 1.44 |
| 2570 | 2.21 ± 0.71 | 3.23 ± 0.73 | 1.99 ± 0.67 | 13.78 ± 2.93 | 6.38 ± 1.33 |
| 2860 | 1.74 ± 0.55 | 2.97 ± 0.66 | 2.65 ± 0.64 | 10.79 ± 2.33 | 5.34 ± 1.10 |
| 3430 | 1.49 ± 0.44 | 3.01 ± 0.60 | 1.87 ± 0.41 | 11.66 ± 2.48 | 5.89 ± 1.03 |
| 3710 | 1.23 ± 0.39 | 2.87 ± 0.55 | 2.10 ± 0.47 | 10.80 ± 2.04 | 5.73 ± 1.03 |

$$p = \exp\left(\frac{-2\pi H_{ij}^2}{\Delta F v_{\text{rad}}}\right), \quad (9)$$

where the transition matrix element H_{ij} is one-half of the splitting of the adiabatic potential-energy curves at the crossing point R_c . ΔF is the difference in the slopes of the corresponding potential-energy curves at R_c and v_{rad} the radial velocity at R_c . The crossing points are calculated by assuming repulsive Coulomb interaction for the $N^{3+}(3\ell)$ - H_2 system and ion-induced dipole interaction for the N^{4+} - H_2 system. Furthermore straight line trajectories are assumed. To obtain the coupling elements H_{ij} the Olson-Salop-Taulbjerg approximation [17–19] is used, which is given by

$$H_{ij} = \left(\frac{9.13f_{nl}}{\sqrt{q}}\right) \exp\left(\frac{-1.324R_c\alpha}{\sqrt{q}}\right). \quad (10)$$

This formula for the matrix elements is based on an empirical fit to exact calculations of the splitting of adiabatic potential curves. The addition of the factor f_{nl} to the original formula of Olsen and Salop allows for describing capture into the nondegenerate ℓ states of partially stripped ions [19]. For the $n=3$ level f_{3l} is equal to 0.58, -0.71 , and 0.41 for $3s$, $3p$, and $3d$, respectively. The possibility to calculate capture from different targets is covered by $\alpha = \sqrt{2I_t}$, with I_t the ionization potential of the target. The probability P_{ij} to transfer an electron from state i to state j equals $2p(1-p)$. The generalization to a situation where N states are involved is straightforward and can, for example, be found in [20]. Now the cross section σ_j for capture into state j can be calculated by

$$\sigma_j = 2\pi \int_0^{R_j} P_{ij}(b) b db. \quad (11)$$

The singlet and triplet systems are treated separately. The corresponding cross sections are afterwards multiplied by their spin statistical weights (0.25 for the singlet states and 0.75 for the triplet states) to allow for a direct comparison with the experimental data. Only capture into the $N^{3+}(3\ell)$ states is found to be of relevance.

The results of the Landau-Zener model are in fair agreement with the experimental results (see Fig. 3). Both the magnitude and energy dependence are predicted well for all

the states with a low angular momentum ℓ . However, the cross sections for capture into both the $3d \ ^1D$ and the $3d \ ^3D$ state are not only clearly underestimated but also the experimentally observed primary energy dependence of the $3d \ ^3D$ state is completely opposite as theoretically predicted. This can be attributed to the fact that the Landau-Zener model only calculates capture into the $m=0$ states and does not include rotational coupling. Apparently this coupling is not important for transitions to states with a low angular momentum but becomes more important for states with a high ℓ .

B. One-electron-capture cross sections for N^{4+} colliding on H

The experimental results for electron capture onto N^{4+} ions in collisions with atomic hydrogen are given in Table II and are depicted as a function of energy in Fig. 4. Again as in Table I the relative errors represent the statistical errors at a 90% confidence level and the uncertainties in the target density. The latter ones now additionally include fluctuations in the dissociation degree of the target. The systematic error is 20–25%.

The TES data by McCullough *et al.* [4] resolve states with different angular momentum, ℓ , but do not resolve the

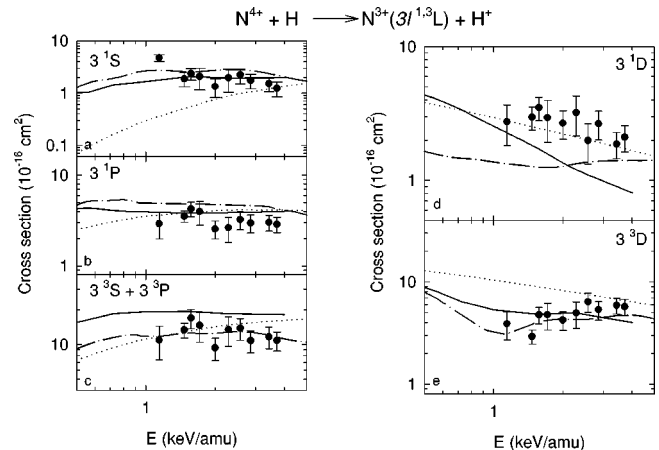


FIG. 4. Comparison of our measured state-selective single-electron-capture cross sections for $N^{4+} + H$ collisions with the theoretical predictions of Stancil *et al.* [1] (---), Shimakura *et al.* [8] (—), and our MCLZ calculation (···).

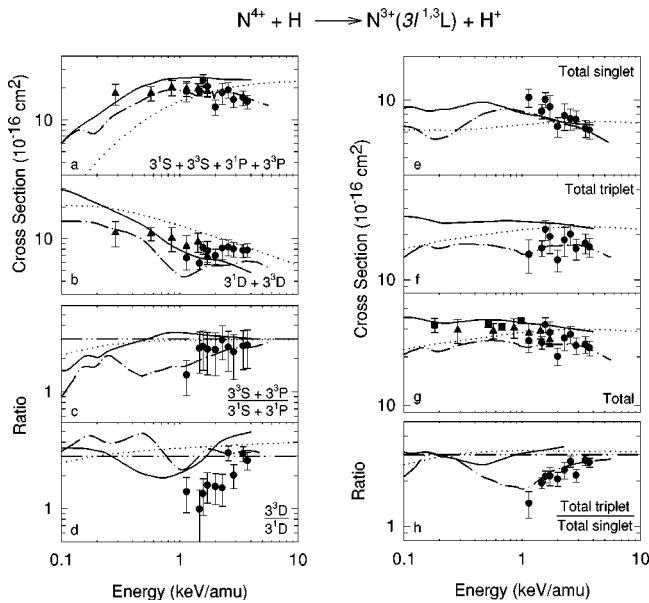


FIG. 5. Comparison of our summed single-electron-capture cross sections and our triplet-singlet ratios (\bullet) for $N^{4+} + H$ with the theoretical predictions of Stancil *et al.* [1] (---), Shimakura *et al.* [8] (—), and our MCLZ calculation (· · ·). The --- lines in panels (c), (d), and (h) indicate a statistical distribution over the two spin states. The other experimental data in panels (a), (b), and (h) are from McCullough *et al.* [4] (\blacktriangle) and Huq *et al.* [2] (\blacksquare).

different spin states. Therefore the sums over both the singlet and triplet states are depicted in Figs. 5(a) and 5(b) to be able to compare our data with their results. There is excellent agreement between the cross sections obtained with both methods in the overlapping energy range. McCullough *et al.* put their cross sections on an absolute scale by normalizing their total cross sections to the merged-beam data of Huq *et al.* [2]. The agreement between our data and the ones of McCullough *et al.* thus implies that our total cross sections should agree with the total cross sections of Huq *et al.* This can indeed be seen from Fig. 5(g).

In a first approach to theoretically analyze N^{4+} -H collisions Zygelman *et al.* [7] calculated only capture into the singlet states. They assumed that the cross sections for capture into the triplet states were equal to the ones of the corresponding singlet states. This implied a statistical distribution over the spin systems. Since the binding energies of the $3l$ states in the triplet system and singlet system are almost equal (cf. Fig. 1) this is a logical assumption. Therefore also the outcome of MCLZ calculations, which mainly depend on the binding energies of the active states, should yield a statistical distribution over the two spin systems. This can indeed be seen from Figs. 5(c), 5(d), and 5(h). From the same figure it is obvious that the most elaborate calculations by Stancil *et al.* [1] and Shimakura *et al.* [8] predict triplet-singlet ratios, which clearly deviate from 3 and which strongly depend on collision energy. Stancil *et al.* used a fully quantum-mechanical close-coupled, molecular-orbital method to determine the cross sections [1]. The molecular data are *ab initio* obtained using a spin-coupled valence-bond method. For the calculation of the coupling-matrix elements four $^1\Sigma^+$ and five $^3\Sigma^+$ states are included. Shimakura *et al.* used a semiclassical close-coupling molecular-

orbital method including electron translation factors. Two-electron processes, i.e., capture and simultaneous projectile-core excitation, have also been considered but over the whole energy region their contribution is small. The molecular electronic states were obtained by using a modified valence-bond configuration-interaction method. In the calculation for the singlet system all eight Σ states and five Π states have been included while on the triplet side all 7 Σ states and five Π states have been taken into account.

A deviation from a statistical distribution is also observed in our measurements [Figs. 5(c), 5(d), and 5(h)]. This triplet-singlet ratio can reliably be determined because all the emission lines are measured in one spectrum, so possible errors due to target fluctuations and changes in beam overlaps cancel. The uncertainty in the ratio is just due to statistical errors and the uncertainty in the relative calibration of the VUV spectrometer, which over the relevant spectral range of 24–39 nm is $\leq 15\%$. From Fig. 5(h) it is seen the ratio of capture into the singlet and triplet states is best described by the calculations of Stancil *et al.* [1]. The difference between the calculations of Stancil *et al.* and Shimakura *et al.* [8] stems mainly from the differences in the ratios for the $3s$ and $3p$ states. From Fig. 4 and in particular Fig. 4(c) it is seen that the main difference in the calculations is in the triplet $3s$ and $3p$ cross sections. Although we realize that there is a systematic error of about 25% associated with our experiments, it seems that for capture into the $3p$ ^3S+3p 3P states the results of Shimakura *et al.* are slightly too high. Furthermore for capture into the $3d$ 1D [Fig. 4(d)] we find a difference between experiment and theory, either in absolute value or in energy dependence. For the other states the differences between the theoretical predictions are too small to distinguish between both models.

IV. CONCLUSION

Cross sections for N^{4+} colliding on atomic hydrogen have been measured in the energy region between 1140 and 3710 eV/amu in which the singlet and triplet states have been resolved. The capture cross sections are not statistically distributed over the singlet and triplet states, agreeing with large-scale calculations by Stancil *et al.* [1] and Shimakura *et al.* [8]. In the energy range of 1 – 4 keV/amu, it is hard to discriminate between the two model calculations even on the basis of state-selective cross sections, because the differences between the two models are too small. From the measured singlet-triplet ratio it is seen that the experimental data are best reproduced by the calculations of Stancil *et al.*

ACKNOWLEDGMENTS

We would like to thank B. Zygelman for providing his data prior to publication and N. Shimakura for providing us with an electronic version of his results. Furthermore, we would like to acknowledge the excellent support of the technical staff of the Kernfysisch Versneller Instituut. This work is part of the research program of the ‘‘Stichting voor Fundamenteel Onderzoek der Materie’’ (FOM), with financial support from the ‘‘Nederlandse Organisatie voor Wetenschappelijk Onderzoek’’ (NWO) and EURATOM.

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