

State-selective one-electron capture by slow state-prepared $\text{N}^{2+}({}^2\text{P})$ ground-state ions in collisions with hydrogen atoms

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Abstract

The technique of double translational energy spectroscopy (DTES), recently successfully developed in this laboratory for use with targets of atomic hydrogen, has been used to study one-electron capture by ground-state $\text{N}^{2+}(2s^2 2p)^2 \text{P}^0$ ions in collisions with hydrogen atoms at energies within the range 0.8–6.0 keV. Cross sections for the formation of the main excited product channels have been determined. The measurements allow a re-evaluation of our previous TES measurements carried out with N^{2+} primary beams containing an admixture of metastable $\text{N}^{2+}(2s 2p^2)^4 \text{P}$ ions. The main findings of these earlier measurements are confirmed and the DTES measurements now remove any ambiguity in interpretation of the experimental data. While recent theoretical studies correctly predict the two main $\text{N}^+ {}^3\text{D}^0$ and ${}^3\text{P}^0$ product channels, the quantitative agreement with experiment is only partially satisfactory.

1. Introduction

One-electron capture collisions of the type



involving collisions between partially ionized species and hydrogen atoms have direct relevance to both astrophysical situations (cf Cravens 1997) and fusion devices (cf Janev 1993). It is well known that, at low velocities $v < 1$ au, a limited number of exothermic channels leading to product states n, l may dominate the electron capture process as a result of pseudo-crossings between the initial and final potential energy curves of the molecular systems formed during the collision.

Accurate theoretical descriptions of these processes are difficult and have relied greatly on experimental measurements to establish the range of validity of the various approximations used. For many years, both in this and other laboratories, the well established technique of translational energy spectroscopy (TES) has been used to identify and determine the relative importance of the collision product channels. In this technique, we carry out careful measurements of the difference ΔT between the kinetic energy of the X^{q+} primary ions and

the forward-scattered X product ions. This allows an assessment of the relative yields of the collision products arising from channels characterized by the energy defects ΔE since, under typical experimental conditions, $\Delta T \approx \Delta E$ (cf McCullough *et al* 1984). However, in many previous measurements a major difficulty of interpretation has arisen through the fact that the observed energy change spectra contain channels associated with unknown fractions of metastable ions as well as ground-state ions in the primary ion beam. Thus, while the TES approach can often identify the presence of product channels arising from metastable primary ions, a quantitative analysis of the energy change spectra is precluded unless the metastable content of the beam is known. The latter often depends strongly on the type and conditions of operation of the ion source used and different fractions can also account for substantial discrepancies between total electron capture cross sections measured in different laboratories.

In this laboratory, we started to address this serious problem by use of the technique of double translational energy spectroscopy (DTES) in an apparatus successfully developed by Greenwood *et al* (1996). We have subsequently demonstrated the effectiveness of this approach by carrying out studies of a variety of processes involving stable gas targets (cf the review by Gilbody (1999)). In this approach, we first convert an intense beam of $X^{(q+1)+}$ ions (from an ECR ion source) into the required X^{q+} ions by electron capture collisions in passage through a suitable gas target such as helium. TES is then used to identify and select X^{q+} ions in either ground or metastable states. These selected product ions in a well defined state are then employed as the primary beam in a second target cell to study the collision process of interest. A second stage of TES is used to identify and quantify the collision product channels unambiguously.

Very recently (Voulot *et al* 2000), we successfully adapted our DTES apparatus to carry out measurements in atomic hydrogen for the first time; measurements of one-electron capture in 6 keV C^{2+} -H(1s) collisions were reported. Unlike our many previous TES studies which employed a tungsten tube furnace to provide a target of highly dissociated hydrogen, our DTES apparatus makes use of a specially developed aluminium target cell. This is fed with highly dissociated hydrogen from a microwave-driven discharge source (see the review by McCullough (1997)).

In this paper our DTES apparatus has been further refined and used to carry out studies of the process



at energies within the range 0.8–6.0 keV. These new measurements allow a comparison with theoretical predictions and a re-evaluation of previous TES measurements in atomic hydrogen carried out by Wilkie *et al* (1985) in this laboratory with primary ion beams containing an unknown fraction of metastable $N^{2+}(2s2p^2)^4P$ in addition to ground-state $N^{2+}(2s^22p)^2P^o$ primary ions.

2. Experimental approach

2.1. General description

The basic experimental arrangement of our DTES apparatus has been described in detail previously (Greenwood *et al* 1996) so that only a brief summary need be given here. A beam of state-prepared N^{2+} primary ions was produced as in our earlier work (Burns *et al* 1997) by first extracting a beam of N^{3+} ions from the ECR ion source/accelerator. This was focused and decelerated by an electrostatic lens system prior to analysis by a hemispherical electrostatic analyser where it emerged with an energy of 180 eV. After further focusing and acceleration to

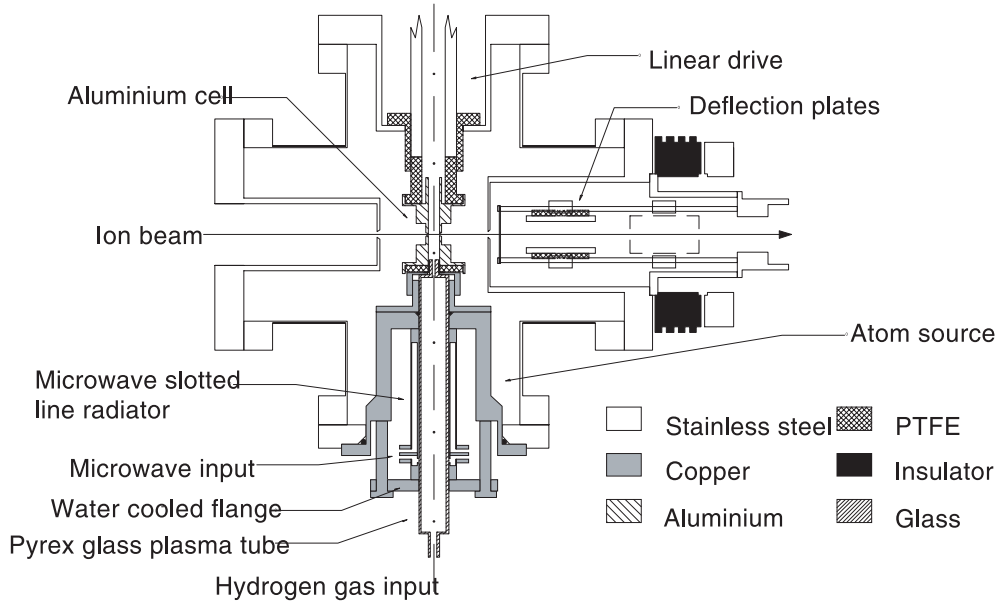


Figure 1. Schematic diagram of the hydrogen atom target comprising the aluminium target cell and microwave-driven hydrogen discharge source.

about 3 keV the resulting beam, with an energy spread of about 1 eV FWHM, passed through the first target cell containing helium. The fast forward-scattered N^{2+} ions formed from the one-electron capture process



were then analysed by a second hemispherical electrostatic analyser. The states of the emergent $N^{2+}(n, l)$ product ions could then be identified by TES through the observed energy changes $\Delta T \approx \Delta E$. As before, in order to prepare a sufficiently intense beam of 100% pure $N^{2+}(2s^2 2p)^2 P^o$ ground-state ions, excited $N^{2+}(2s 2p^2)^2 P$ ions from the main excited product channel were selected. Ions in this state are known to decay rapidly (Wiese *et al* 1966) to the ground state within the minimum transit time (8 μs) of the ions to the atomic hydrogen target. As in our previous work (see figure 1 in Burns *et al* (1997)) the available energy resolution was sufficient to cleanly separate $N^{2+}(2s 2p^2)^2 P$ products from other excited products. It is worth noting that $N^{2+}(2s 2p^2)^4 P$ metastable ions can only be formed by electron capture by $N^{3+} {}^3 P^o$ metastable ions since capture by $N^{3+} {}^1 S$ ground-state ions is a spin-forbidden process. However, as shown previously (see figure 1 in Burns *et al* (1997)), although the N^{3+} beam from the ion source contained both $N^{3+}(1s^2 2s^2) {}^1 S$ ground state and $N^{3+}(1s^2 2s 2p) {}^3 P^o$ metastable ions, none of the observed electron capture channels in helium associated with the $N^{3+} {}^3 P^o$ component were found to result in $N^{2+} {}^4 P$ metastable ion formation. For this reason, the present DTES measurements have had to be confined to the use of pure ground state $N^{2+} {}^2 P^o$ ion beams.

2.2. The hydrogen atom target

A schematic diagram of the hydrogen atom target comprising the aluminium target cell and microwave-driven hydrogen discharge source is shown in figure 1. The target cell, fabricated

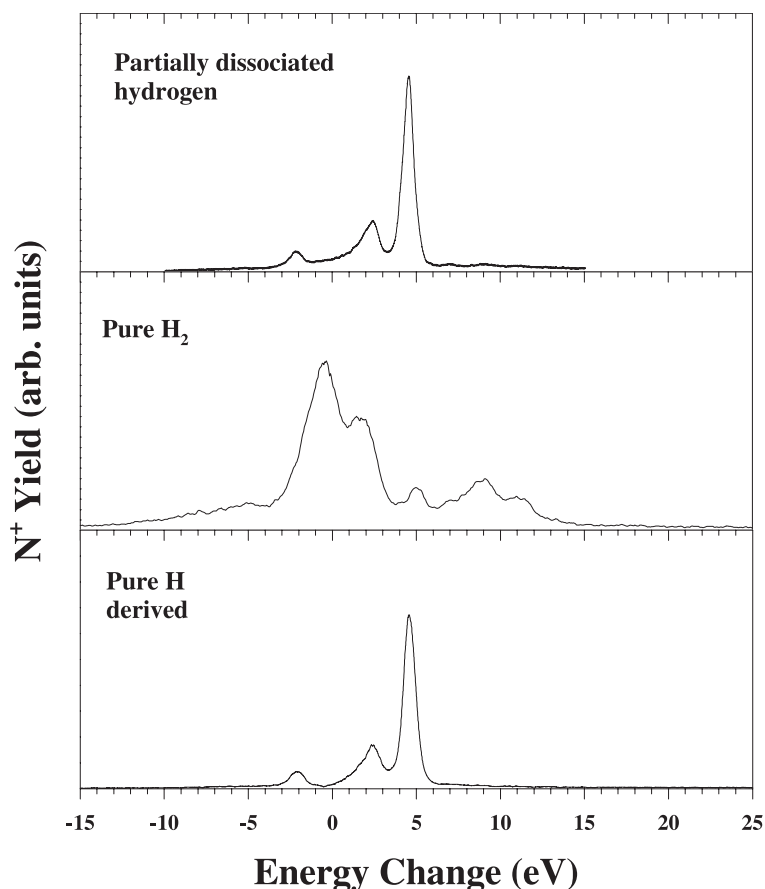


Figure 2. Typical energy change spectra at 3 keV for one-electron capture by pure $N^{2+} 2P^0$ ground-state ions measured in highly dissociated (about 70%) hydrogen, in pure molecular hydrogen and the derived spectrum (see text) for pure atomic hydrogen.

from a 10 mm diameter aluminium tube, was mounted at 90° with respect to the beam direction. The beam passed through a 0.8 mm diameter entrance aperture in the middle of the tube and emerged through a 1.2 mm exit aperture diametrically opposite. The tube was mounted on PTFE insulators and held typically at a potential of +50 V with respect to the surrounding region. This voltage labelling ensured that the collisions occurring in the region outside the cell did not contribute to the observed energy change spectrum.

The dissociation fraction of the hydrogen within the aluminium target cell could be assessed by observing the fractional reduction in one of the peaks in the pure H_2 energy change spectrum (in the region $\Delta T > 6$ eV where no contributions from H were present) when the microwave discharge source was extinguished. Thus if S_1 is the signal obtained with the discharge on and S_2 with the discharge off, the dissociation fraction $D = (S_2 - S_1)/S_2$.

In figure 2, typical energy change spectra obtained at 3 keV for one-electron capture by pure $N^{2+} 2P^0$ ground-state ions in highly dissociated (about 70%) hydrogen and pure molecular hydrogen can be seen to be very different; a detailed discussion of the latter has been given in our previous work (Burns *et al* 1997). The corresponding spectrum shown for pure atomic hydrogen was derived by careful subtraction of an appropriate fraction of the

molecular contribution from the spectrum obtained in the highly dissociated hydrogen target. The accurate subtraction process was facilitated by the ability of the hydrogen source to be rapidly switched from highly dissociated hydrogen to pure H_2 . All the contributions to the partially dissociated spectrum above 6 eV could be ascribed to electron capture channels in H_2 . Thus, in this case, an appropriate fraction of the contributions in the entire pure H_2 spectrum (according to the dissociation fraction) was subtracted from the partially dissociated spectrum to reduce the contributions above 6 eV in the latter to zero.

3. Results and discussion

Figure 3 shows energy change spectra obtained for one-electron capture by 6 keV N^{2+} ions in atomic hydrogen. Here the ‘mixed’ beam spectrum for N^{2+} ions obtained directly from the ion source containing an unknown fraction of metastable ions is compared with the spectrum obtained using a state-prepared pure $N^{2+} \ ^2P$ ground-state beam. The positions of the possible product channels corresponding to ground-state primary ions (G) and metastable primary ions (M) are also shown on these spectra. The mixed beam and pure ground state spectra can be seen to be very similar, evidently because collision product channels with not greatly different energy defects provide the dominant contributions to both.

In the case of ground-state primary ions, the main processes can be identified as:

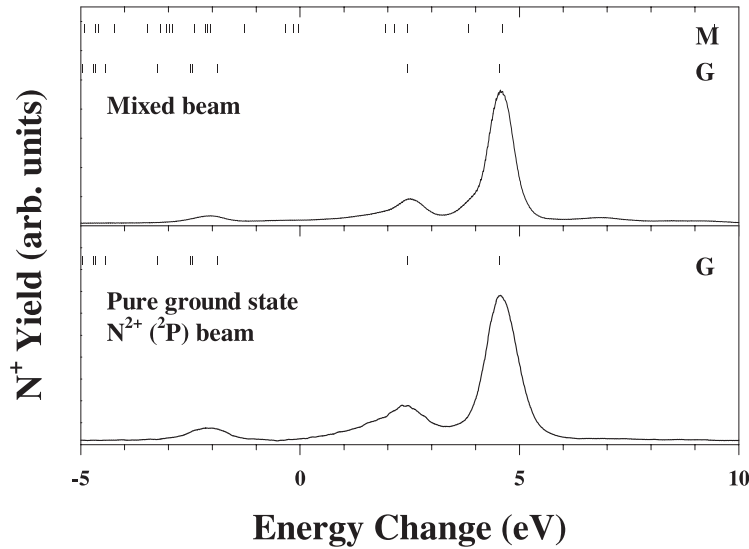
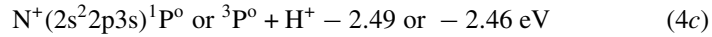
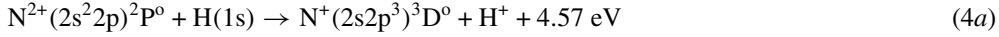
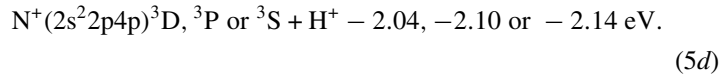
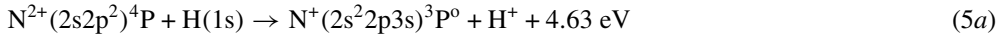


Figure 3. Energy change spectra obtained for one-electron capture by 6 keV N^{2+} ions in atomic hydrogen. The ‘mixed’ beam spectrum for N^{2+} ions obtained directly from the ion source containing an unknown fraction of metastable ions is compared with the spectrum obtained using a state-prepared pure $N^{2+} \ ^2P^0$ ground-state beam. Positions of possible product channels corresponding to ground-state primary ions (G) and metastable primary ions (M) are also shown.

of which the two exothermic channels (4a) and (4b) are dominant; both of these involve core excitation. Our energy resolution can be seen to be insufficient to separate the small $^1\text{P}^0$, $^3\text{P}^0$ and $^1\text{D}^0$ product yields which appear to arise from the endothermic channels (4c) and (4d).

Although analysis of the ‘mixed’ beam spectrum is less clear, careful inspection indicates that the metastable $\text{N}^{2+} 4\text{P}$ component may be associated with the following product channels:



In this paper, the close similarity between the ‘mixed’ beam and pure ground-state energy change spectra confirms the results of our earlier TES measurements carried out in this laboratory (Wilkie *et al* 1985) in the range 0.6–8.0 keV using N^{2+} beams which contained an admixture of metastable ions. In those measurements, changes in the metastable fraction were found to have no detectable effect on the measured total electron capture cross sections, leading us to conclude that the total cross sections for ground and metastable ions N^{2+} in atomic hydrogen were not greatly different. In contrast, the corresponding cross sections in molecular hydrogen were found to be strongly dependent on the metastable fraction present in the primary beam, a result which our DTES measurements (Burns *et al* 1997) have since confirmed in detail.

In figure 4 we show energy change spectra for one-electron capture by pure ground state $\text{N}^{2+} 2\text{P}^0$ ion beams in atomic hydrogen at 6.0, 3.0, 1.5, 1.2 and 0.8 keV. As the impact energy is increased, while the exothermic $\text{N}^+ 3\text{D}^0$ product channel (4a) remains dominant, the less exothermic $\text{N}^+ 3\text{P}^0$ product channel (4b) increases in relative importance. The unresolved $\text{N}^+(^1\text{P}^0 + ^3\text{P}^0 + ^1\text{D}^0)$ contributions from the endothermic product channels in (4c) and (4d) can be seen to only become significant at the highest energies considered. In order to obtain cross sections for the individual product channels from these energy change spectra, we have normalized the data to our previously measured total electron capture cross sections (Wilkie *et al* 1985). The latter values are in good agreement with measurements by Seim *et al* (1981) and Phaneuf *et al* (1978) in the energy ranges of overlap. This normalization procedure seems justifiable in view of the lack of dependence of these total cross sections on the metastable content of the primary beam.

Table 1 shows the cross sections for the individual product channels derived from the present DTES measurements. The quoted uncertainties reflect the reproducibility of the

Table 1. Cross sections for formation of $\text{N}^+ 3\text{D}^0$ through (4a), $^3\text{P}^0$ through (4b) and $\text{N}^+(^1\text{P}^0 + ^3\text{P}^0 + ^1\text{D}^0)$ through (4c) and (4d) in $\text{N}^{2+} 2\text{P}^0\text{--H}(1s)$ collisions.

Energy (keV)	Cross sections (10^{-16} cm^2)		
	$\text{N}^+ 3\text{D}^0$ through (4a)	$\text{N}^+ 3\text{P}^0$ through (4b)	$\text{N}^+(^1\text{P}^0 + ^3\text{P}^0 + ^1\text{D}^0)$ through (4c) and (4d)
0.8	3.19 ± 0.26	0.32 ± 0.26	—
1.2	2.97 ± 0.36	0.40 ± 0.37	—
1.5	3.16 ± 0.31	0.30 ± 0.31	—
3.0	3.80 ± 0.63	0.88 ± 0.51	0.15 ± 0.68
6.0	3.53 ± 0.53	1.70 ± 0.45	0.43 ± 0.62

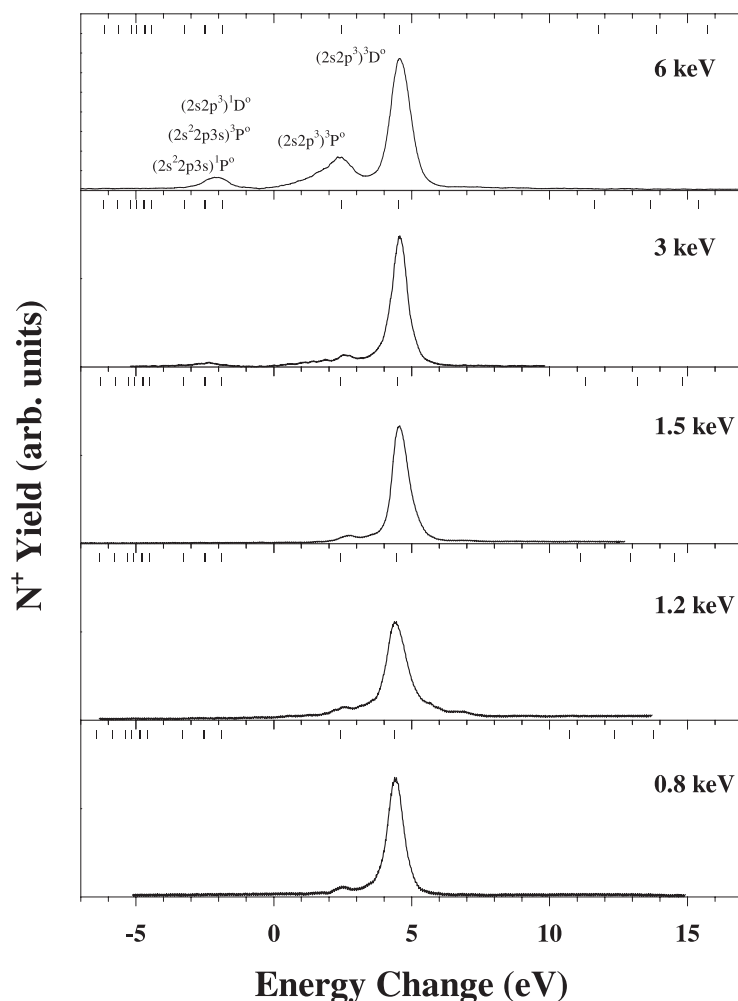


Figure 4. Energy change spectra for 6, 3, 1.5, 1.2 and 0.8 keV pure $N^{2+} 2P^0$ ground-state ions in atomic hydrogen. Positions of possible product channels are also shown.

measurements shown as 95% confidence limits. In figure 5, these cross sections may be compared with the corresponding cross sections obtained in our previous TES measurements. The general agreement between the two sets of data can be seen to be very satisfactory for the $^3D^0$, $^3P^0$ and $N^+(^1P^0 + ^3P^0 + ^1D^0)$ product channels. It is important to note Wilkie *et al* (1985), in their lower-resolution measurements, identified the small peak comprising the $N^+(^1P^0 + ^3P^0 + ^1D^0)$ contributions from endothermic channels as due to the $N^+(^1D^0)$ product channel alone. In addition, Wilkie *et al* (1985) also recorded at and above 6 keV very small contributions from an $N^+(2s^22p3p)^3P$ product channel corresponding to $\Delta E = -5.17$ eV. There is no evidence of any significant yield from this channel in the present higher-resolution DTES measurements.

In figure 5 we include cross sections for the main $^3D^0$ product channel calculated by Bienstock *et al* (1986) using a molecular approach; this is an extension of earlier theoretical work by Heil *et al* (1981). These calculations predict the $^3D^0$ product channel is the only

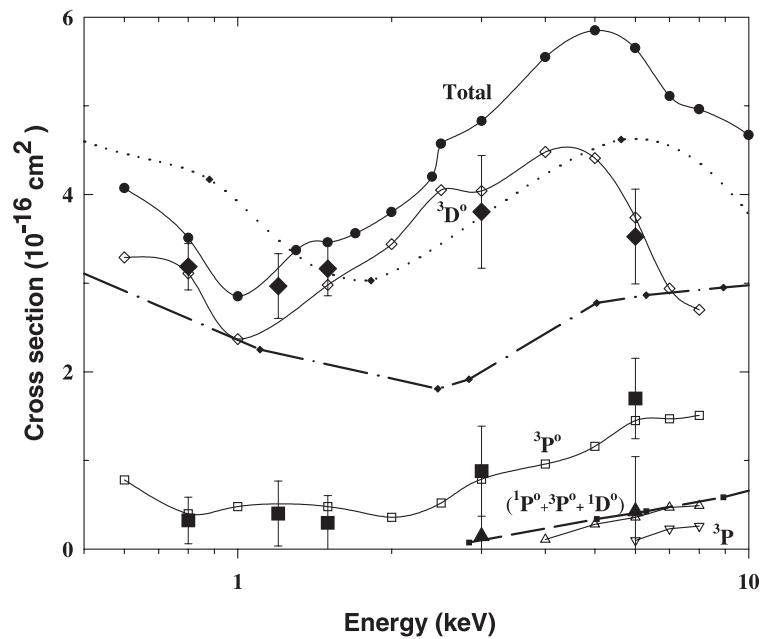


Figure 5. Cross sections for one-electron capture by N^{2+} ions in atomic hydrogen leading to specified $N^+(n, l)$ products. Present DTES measurements with pure ground-state $N^{2+} 2P^0$ ions: \blacklozenge , $^3D^0$; \blacksquare , $^3P^0$; \blacktriangle , $(^1D^0 + ^1P^0 + ^3P^0)$. Our previous TES measurements (Wilkie *et al* 1985) using an N^{2+} beam containing an unknown fraction of metastable ions: \bullet , total; \diamond , $^3D^0$; \square , $^3P^0$; \triangle , $(^3P^0 + ^1P^0 + ^1D^0)$; ∇ , 3P ; theory by Bienstock *et al* (1986): \cdots , $^3D^0$. Theory by Herrero *et al* (1995): $-\cdot-$, $^3D^0$; $---$, $^3P^0$.

significant electron capture channel at the energies considered and are therefore at variance with our experimental findings. However, since the $^3D^0$ channel provides the dominant contribution, the calculated values can be seen to be in reasonable accord with our experimental values.

More recent quantal calculations have been carried out by Herrero *et al* (1995) in which molecular states were obtained using *ab initio* SCF-CI methods. These calculations, which extend to much lower energies than we consider, correctly predict contributions from channels other than the dominant $^3D^0$ product channel. As indicated in table 1, in the present energy range, Herrero *et al* (1995) predict fractional contributions from both the $^3D^0$ and $^3P^0$ product channels which are in rough general accord with experiment. However, the calculated cross sections (figure 5) for these channels can be seen to be in less satisfactory agreement with the experimental values.

Table 2. Percentages of the total yield for the main product channels arising from one-electron capture by $N^{2+} 2P^0$ ground-state ions in atomic hydrogen. Values (E) obtained from the present experiment are compared with theoretical values (T) predicted by Herrero *et al* (1995).

N^+ product	6.0 keV		3.0 keV		1.5 keV		1.2 keV		0.8 keV	
	E	T	E	T	E	T	E	T	E	T
$^3D^0$ through (4a)	78.0	86.7	95.2	93.0	97.7	100	81.1	100	90.5	100
$^3P^0$ through (4b)	15.2	13.3	2.8	7.0	2.3	—	18.9	—	9.5	—
$(^1P^0 + ^3P^0 + ^1D^0)$ through (4c) and (4d)	6.8	—	2.1	—	—	—	—	—	—	—

4. Conclusion

The technique of DTES, only recently applied successfully to targets of atomic hydrogen, has now been used to study one-electron capture in $N^{2+} 2P^0-H(1s)$ collisions at energies in the range 0.8–6 keV and obtain cross sections for the main excited product channels. In view of the fact that previous measurements have been carried out with ion beams containing an unknown admixture of metastable $N^{2+}(2s2p^2)^4P$ in addition to ground-state $N^{2+}(2s^22p)^2P^0$ ions, it was of particular interest to re-evaluate our earlier TES measurements (Wilkie *et al* 1985). The present DTES confirm the main findings of these earlier measurements and remove any uncertainty in interpretation of the experimental data. At the energies considered, while the two main $N^+ 3D^0$ and $3P^0$ product channels observed experimentally are correctly predicted by current theory, the quantitative agreement with the calculations is only partially satisfactory.

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References

- Bienstock S, Dalgarno A and Heil T G 1986 *Phys. Rev. A* **33** 2078
Burns D, Greenwood J B, McCullough R W, Geddes J and Gilbody H B 1997 *J. Phys. B: At. Mol. Opt. Phys.* **30** L323
Cravens T E 1997 *Geophys. Res. Lett.* **24** 105
Gilbody H B 1999 *Nucl. Instrum. Methods B* **154** 113
Greenwood J B, Burns D, McCullough R W, Geddes J and Gilbody H B 1996 *J. Phys. B: At. Mol. Opt. Phys.* **29** 5867
Heil T G, Butler S E and Dalgarno A 1981 *Phys. Rev. A* **23** 1100
Herrero B, Cooper I L, Dickinson A S and Flower D R 1995 *J. Phys. B: At. Mol. Opt. Phys.* **28** 711
Janev R K 1993 *IAEA Technical Committee Meeting on Atomic and Molecular Data for Fusion Reactor Technology (Cadarahe, France), IAEA Report INDC (NDS)-277* (Vienna)
McCullough R W 1997 *Proc. 14th Int. Conf. on the Application of Accelerators in Research and Industry (Denton, OH) (AIP Conf. Series no 392)* (New York: AIP) p 275
McCullough R W, Lennon M, Wilkie F G and Gilbody H B 1984 *J. Phys. B: At. Mol. Phys.* **17** 1373
Phaneuf R A, Meyer F W and McKnight R A 1978 *Phys. Rev. A* **17** 534
Seim W, Muller A, Wirkner-Bott I and Salzborn E 1981 *J. Phys. B: At. Mol. Phys.* **14** 3475
Voulot D, Gillen D R, Thompson W R, Gilbody H B, McCullough R W, Errea L, Macias A, Mendez L and Riera A 2000 *J. Phys. B: At. Mol. Opt. Phys.* **33** L187
Wiese W L, Smith M W and Glennon B M 1966 *Atomic Transition Probabilities* vol 1 *National Standard Reference Data Series 4* (Washington, DC: US Govt Printing Office)
Wilkie F G, Yousif F B, McCullough R W, Geddes J and Gilbody H B 1985 *J. Phys. B: At. Mol. Phys.* **18** 479