Low-energy electron capture by N^{2+} ions from atomic hydrogen using merged beams

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Using the merged-beams technique, independent, absolute, total electron-capture cross sections have been measured for the $N^{2+}+H$ (or D) system in the collision energy range 0.1–4500 eV/amu. For collision energies less than 200 eV/amu, the data are in reasonable accord with recent fully quantal calculations of Herrero *et al.* [J. Phys. B **28**, 711 (1995)], except that the experiment provides no evidence for the existence of a peak in the cross section at 0.25 eV/amu. Additional structure between 10 and 100 eV/amu, as predicted by Bienstock *et al.* [Phys. Rev. A **33**, 2078 (1986)], is not observed either. A comparison is also made with other measurements, which make use of a hydrogen furnace [Phaneuf *et al.*, Phys. Rev. A **17**, 534 (1978); Seim *et al.*, J. Phys. B **14**, 3475 (1981); Wilkie *et al.*, J. Phys. B **18**, 479 (1985)]. Above 2000 eV/amu the agreement is excellent. However, between 200 and 2000 eV/amu the present results are some 30% below the previously published experimental and theoretical cross sections, and a local maximum is found near 200 eV/amu, instead of 360 eV/amu. A possible explanation for this discrepancy is suggested. [S1050-2947(97)07104-7]

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I. INTRODUCTION

Many current investigations point to the practical relevance of electron-capture processes in astrophysics and plasma research, in particular fusion reactor design, where nowadays the emphasis is on atomic collisions in the divertor region [1]. Especially at low collision energies, there are also fundamental physics issues that have yet to be resolved. At such low energies, the collision process possesses strong molecular features, which yields stringent tests for theoretical methods. Just recently, very accurate theoretical calculations of the electron-capture cross sections for Si^{4+} ions impacting on H and D atoms at thermal and near-thermal collision energies helped to demonstrate that trajectory effects caused by the ion-induced dipole attraction lead to a $1/v$ scaling, v being the collision velocity, and a kinematic isotope effect (see Ref. [2] and references therein for a more comprehensive discussion). Orbiting or shape resonances in the electron-capture cross section for multicharged ions in collisions with H at thermal energies have been predicted by fully quantum mechanical calculations [3], but yet await experimental confirmation.

The work presented here focuses on the following process:

$$N^{2+}(1s^2 2s^2 2p,{}^2P^{\circ})+H(1s)\rightarrow N^{+}+H^{+}, \qquad (1)$$

where the final state of N^+ is not specified, i.e., the sum of all partial electron-capture channels is considered. The main motivation for this study is to verify experimentally the predicted (oscillatory) structure in the total electron-capture cross section at low energies [4,5]. Besides, as was discussed by Herrero *et al.* [5], production of N^+ is also of astrophysical importance, because forbidden lines of this ion have been studied extensively for many years in gaseous nebulae. For practical reasons, in the experiment both atomic hydrogen and its isotope deuterium were used as target gases.

Below a collision energy of 100 eV/amu, it has been established experimentally as well as theoretically that the dominant channel for process 1 is capture into the $N^+(^3D^o)$ state [5,6]. At higher energies, the $N^+(^3P^o)$ channel, in particular, but also the $N^+(^1D^o)$ and $N^+(^3P)$ channels, start to contribute to the total cross section [6]. Capture into the $^3P^o$ and $^3D^o$ states of N^+ are processes exothermic by 2.18 and 4.29 eV, respectively [5,7].

A state-of-the-art merged-beams technique provides an accurate tool for studying electron capture in a broad region of collision energies, ranging from the thermal to the keV scale. The experimental setup and technique will be briefly described. The presently obtained data will be compared with other experimental [6,8,9] and theoretical [4,5] results, and a possible explanation will be given for observed discrepancies.

II. EXPERIMENTAL TECHNIQUE

Measurements of the total electron-capture cross section for $N^{2+}+H(D)$ were carried out with the Oak Ridge National Laboratory (ORNL) ion-atom merged-beams apparatus. A detailed description of the apparatus and the techniques involved can be found in the literature [10]. Only the essential features will be discussed here. A schematic representation of the setup is shown in Fig. 1.

Collisions between multicharged ions and atomic hydrogen (deuterium) in a merged-beams configuration provide a large dynamic range of collision energies, accessing energies below 10 eV/amu, where more straightforward methods using a hydrogen furnace [6,8,9] fail. In the present experimental undertaking, a 16–50-keV N^{2+} beam was merged with either a 7.25–10-keV D beam or a 10-keV H beam allowing electron-capture cross sections to be measured in the energy range of 0.1–5000 eV/amu. The center-of-mass collision energy (eV/amu) is given by

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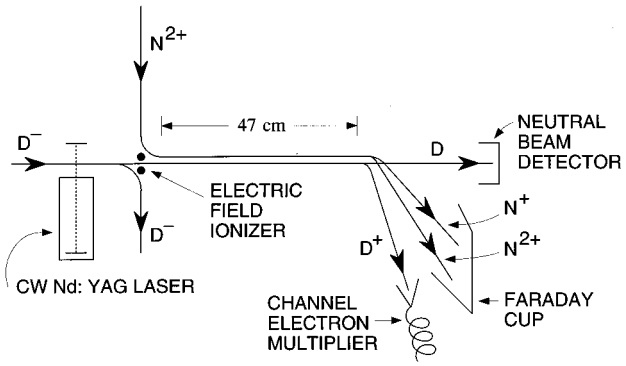

FIG. 1. Schematic drawing of the merged-beams setup.

$$E_{\text{c.m.}}=\left[\frac{E_1}{m_1}+\frac{E_2}{m_2}\right]-2\sqrt{\frac{E_1E_2}{m_1m_2}}\cos(\theta), \tag{2}$$

where E_1 and m_1 refer to the laboratory frame energy (eV) and the mass (a.u.) of the neutral beam, and E_2 and m_2 to those of the ion beam. Included in E_1 and E_2 are the estimated plasma potential shifts of the duoplasmatron source (10 V) and the ECR source (20 V per charge) [11]. In most cases, the merge angle θ is very small, but it has to be considered at thermal collision energies, where it puts a limit to the lowest center-of-mass energy that can be reached [2].

Usually, the neutral beam is the fastest one, since the practical acceleration voltage of the ECR source is limited to 25 kV, and the ions are relatively heavy. Consequently, as is generally the case, a deuterium beam is used to access low collision energies, while an atomic hydrogen beam is used to access energies in the keV/amu range. In the region where both data sets connect (around 1500 eV/amu), the data taken with either H or D, which correspond to very different experimental conditions, were found to be consistent. This is an indication that problems with the signal collection efficiency of the present merged-beams setup have been avoided. In other words, there is no evidence of a substantial amount of trajectories of signal ions not reaching the detector. Furthermore, for a typical test case such as $O^{5+}+D$, for which it is known that relatively large angular scattering occurs [12], it has been ascertained, using the same experimental setup, that good agreement exists between theory and experiment [13]. In addition, the merged-beams technique intrinsically favors a large angular acceptance, because the transformation from the center-of-mass to the laboratory frame reduces all scattering angles. Again considering the $O^{5+}+D$ system, with an estimated angular acceptance (in the laboratory frame) of 2.3° [13], it was calculated that all of the signal would be collected below 1 eV/amu, even if 90° scattering would occur in the center-of-mass frame [13]. As the collision energy is increased, large angular scattering in the center-of-mass frame simply becomes more and more unlikely. Based on these arguments, it is concluded that the current angular collection efficiency suffices.

The N^{2+} beam is produced by the ORNL CAPRICE electron cyclotron resonance (ECR) ion source. The beam specifications are an intensity of 1–8 μA, a diameter of 2–4 mm full width at half maximum, and a divergence less than 0.25°. A fast neutral H(D) atom beam is obtained by photodetachment of a keV $H^-(D^-)$ beam extracted from a duoplasmatron source and passed through the resonant cavity of a 1.06 μm cw neodymium-doped yttrium aluminum garnet laser. The H(D) beam is nearly parallel (the divergence is less than 0.15°), has a diameter of about 2 mm, and an intensity of 10–80 nA.

The ion and neutral beams are merged electrostatically. Both beams interact along a field-free region of 47 cm (see Fig. 1), after which the primary beams and the product H^+ (D^+) ions are separated magnetically. Secondary emission from a stainless steel plate is used to monitor the neutral beam intensity, while the intensity of the N^{2+} beam is measured using a Faraday cup. The $H^+(D^+)$ product ions are detected by a channel electron multiplier. The absolute, total electron-capture cross section is obtained from parameters that can all be determined experimentally [10]:

$$\sigma=\frac{Sqe^2\gamma v_1v_2}{I_1I_2\varepsilon\langle F\rangle Lv_r}, \tag{3}$$

where S is the signal count rate, q the initial charge of the ion, e is the electronic charge, γ is the secondary electron emission coefficient of the neutral beam detector, v_1 and v_2 are the velocities of the beams, I_1 and I_2 are the electrical beam intensities, ε is the efficiency for detecting $H^+(D^+)$, $\langle F\rangle$ is the average beam overlap integral, L is the length of the merge path, and $v_r=|v_1-v_2|$ is the relative velocity between beams. The average beam overlap integral was obtained from two-dimensional scans of the beams at three different positions along the merge path [10]. The numerical value for γ was determined *in situ* [10].

Both the neutral and the ion beam contain unwanted constituents that may contribute to the measured signal. Specifically, this concerns H(D) atoms in Rydberg states, resulting from stripping of $H^-(D^-)$ ions with background gas, and N^{2+} metastables. As shown in Fig. 1, the neutral beam passes through an electric field (30 kV/cm) ionizing excited H(D) atoms whose electrons are in high-n shells (typically $n\geqslant 12$), and radiatively quenching atoms with electrons occupying $n<8$ shells, thereby reducing the correction [14] to the cross section to less than 10%.

The amount of N^{2+} metastables in the ion beam was checked in a separate measurement. When the N^{2+} beam was directed into the ORNL electron-ion crossed-beams apparatus [15,16] to look for electron-impact ionization below the ground-state threshold, no signal was detected. The presence of metastable ions could, therefore, be ruled out.

III. RESULTS AND DISCUSSION

In Fig. 2 the measured electron-capture cross section of $N^{2+}+H(D)$ is shown as a function of collision energy. Data were taken during different sessions spanning a period of about one year, and were found to be reproducible. The error bars on the current experimental cross sections, as shown in Fig. 2, indicate the statistical error at a 90% confidence level. The total uncertainty is the quadrature sum of the statistical error and the systematic error, the latter being 12% according to Ref. [14]. The data, including the total uncertainty, are also presented in Table I.

For the lowest energy point in Fig. 2 and Table I, the total

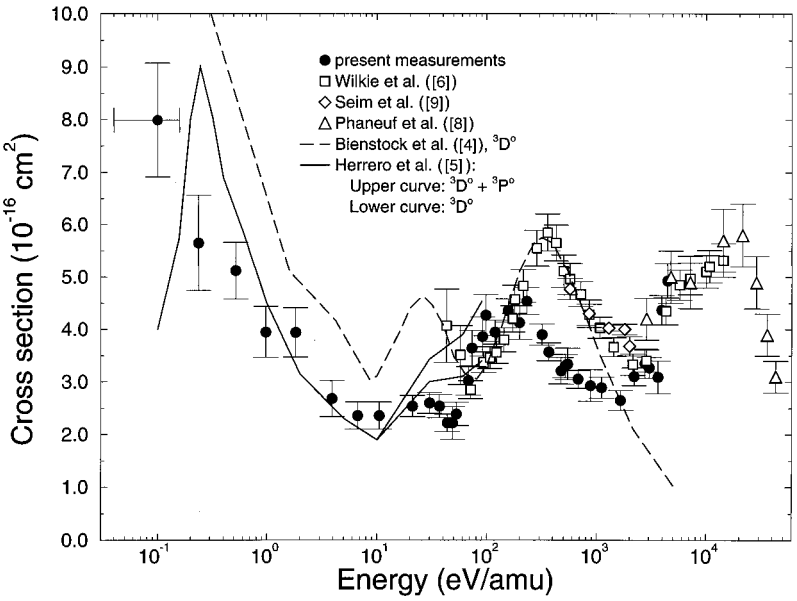

FIG. 2. Dependence of the total electron-capture cross section of $N^{2+}+H(D)$ on the collision energy. The different experimental and theoretical results are denoted in the legend. The lower solid curve is the result of calculations of Herrero *et al.* [5] for capture into the $^3D^o$ state of N^+. The upper solid curve also includes capture into the $^3P^o$ state. The dashed line represents calculations of Bienstock *et al.* [4], which are again only for the $N^+(^3D^o)$ channel.

uncertainty in collision energy is indicated. At this energy, the merge angle between the ion and neutral beams was 0.18°, while the divergence of both beams was less than 0.15°. The collision energy uncertainty is determined by the divergences of the beams, the rms voltage fluctuation of the ECR power supply, and the plasma potential spreads of the ECR and duoplasmatron sources [2].

Also shown in Fig. 2 are other experimental and theoretical results. These include measurements of Phaneuf *et al.* [8], Seim *et al.* [9], and Wilkie *et al.* [6], who all used a hydrogen furnace target, and calculations of Bienstock *et al.* [4], who applied a combination of close-coupled and unitarized distorted-wave techniques, and Herrero *et al.* [5], whose method is fully quantal. The results of Bienstock *et al.* [4] are for capture into the $^3D^o$ state of N^+ only. The same is true for the lower curve of Herrero *et al.* [5] in Fig. 2, while their upper curve also includes capture into the $^3P^o$ state.

As shown in Fig. 2, at energies above 2000 eV/amu the present measurements are in excellent agreement with those of the other experimental groups [6,8]. Theory is not available at these energies. At lower energies, the situation is far less clear. This will now be discussed in more detail.

The results presented here are believed to be reliable: the small amount of excited H or D atoms in the neutral beam affects the cross sections by less then 10%, and proper corrections have been made [14]; the fraction of metastable N^{2+} ions was found to be negligible; and the angular acceptance of the setup should be sufficient. Wilkie *et al.* [6] also exclude effects from metastable ions. In contrast to our experimental technique, however, both Wilkie *et al.* [6] and Seim *et al.* [9] made use of a tungsten tube furnace to produce a partly ($<$ 95%) dissociated hydrogen target. This could perhaps explain the remarkable discrepancy in Fig. 2 between our data and those of these two groups in the energy range 200–2000 eV/amu, where our measured electron-capture cross sections are about 30% below theirs, and a local maximum in the cross section is found at about 200 eV/amu, rather than at 360 eV/amu. We suggest that in the two experiments just mentioned [6,9] a small fraction of the target beam from the hydrogen furnace consisted of excited states, e.g., vibrationally excited H_2, leading to an apparent cross section that is overestimated. For a temperature of 2000 K, at which hydrogen furnaces are typically operated (1950 K for Seim *et al.* [9] and 2500 K for Wilkie *et al.* [6]), the Boltzmann distribution predicts a population of 5% for the first vibrationally excited state ($\nu=1$) [17], but considerable less for higher lying vibrational states. In order to explain the observed discrepancy, electron capture from H_2 ($\nu=1$) by N^{2+} ions would require a cross section 200 times greater than the one for capture from H. In addition, since our results are in reasonable accord again with the data of Wilkie *et al.* [6] below 200 eV/amu, the electron-capture cross section from vibrationally excited hydrogen molecules should quickly decrease outside the 200–2000 eV/amu collision energy window.

The excited state production may also be fed by the chemical process that leads to dissociation and produce (vibrationally) excited states in a variety of levels. For example, in another experiment an effusive molecular beam tungsten furnace, operated at similar temperatures as indicated above, was used to produce $\nu=4,5$ vibrationally excited states of H_2 [18]. A disagreement between results caused by the use of different experimental techniques may only show up in collision systems for which the electron-capture cross sections are relatively small. Indeed, for N^{2+} impact the background contribution of the undissociated H_2 in the oven is significant compared to the signal due to H [19]. Merged-beams measurements for $C^{3+}+H$ between 200 and 3000 eV/amu [11] exhibit a discrepancy resembling the one reported here.

It may only be a coincidence that the *partial* cross section of Wilkie *et al.* [6] for capture into the $N^+(^3D^o)$ state follows our *total* cross section. This means that the discrepancy in Fig. 2 in the 200–2000 eV/amu region corresponds closely to the contribution attributed to capture into the $N^+(^3P^o)$ state [6]. The $N^+(^3P^o)$ channel has a pseudocrossing with the initial channel at an internuclear distance $R \simeq 12.5a_0$ [20]. However, as was discussed by Herrero *et al.*

TABLE I. Ion-atom merged-beams cross section data for $N^{2+}+H(D)$ as a function of collision energy. Also listed are the relative uncertainty and total combined (relative plus systematic) uncertainty estimated at the 90% confidence level. The uncertainty in the collision energy is given at 0.10 eV/amu.

Neutral	Collision energy (eV/amu)	Cross section (10^{-16} cm^2)	Relative uncert. (10^{-16} cm^2)	Total uncert. (10^{-16} cm^2)
D	0.10 ± 0.06	7.99	1.08	1.44
D	0.238	5.65	0.91	1.13
D	0.52	5.12	0.54	0.82
D	0.98	3.95	0.49	0.68
D	1.83	3.94	0.47	0.67
D	3.92	2.68	0.34	0.47
D	6.69	2.36	0.26	0.38
D	10.5	2.36	0.26	0.38
D	21.0	2.54	0.20	0.36
D	30.0	2.60	0.20	0.37
D	36.8	2.54	0.22	0.38
D	43.4	2.22	0.17	0.32
D	48.6	2.22	0.31	0.41
D	52.6	2.39	0.22	0.36
D	67.9	3.02	0.27	0.45
D	73.1	3.64	0.34	0.55
D	91.1	3.86	0.37	0.59
D	97.6	4.27	0.39	0.64
D	118	3.95	0.23	0.53
D	156	4.36	0.22	0.57
D	198	4.13	0.32	0.59
D	230	4.54	0.26	0.60
D	319	3.90	0.20	0.51
D	368	3.57	0.17	0.46
D	474	3.21	0.25	0.46
D	518	3.30	0.22	0.45
D	542	3.34	0.30	0.50
D	677	3.05	0.17	0.40
D	881	2.93	0.30	0.46
D	1107	2.89	0.20	0.40
H	1654	2.65	0.19	0.37
H	2204	3.10	0.17	0.41
H	2770	3.35	0.28	0.49
H	3019	3.27	0.20	0.44
H	3624	3.09	0.31	0.48
H	3929	4.37	0.28	0.59
H	4445	4.93	0.33	0.68

[5], a second pseudocrossing at $R\simeq 2.2a_0$ is believed to be more important for populating $N^+(^3P^o)$. It is unfortunate that at such a small R the potential energy curves are rising exponentially, and therefore the Landau-Zener model (see, e.g., Ref. [21]) cannot be used to estimate the position of the maximum of this partial electron-capture cross section.

Comparing measurements to theory, Wilkie *et al.* [6] agree perfectly with Bienstock *et al.* [4] up to 1000 eV/amu, where capture into the $N^+(^3D^o)$ state is no longer dominant. This perfect agreement is, as admitted by Bienstock *et al.* [4] ''somewhat fortuitous,'' since a comparison between their results and the partial cross section for capture into the $N^+(^3D^o)$ state, as measured by Wilkie *et al.* [6], shows that the calculated cross section is actually too big. As can be seen from Fig. 2, our total cross sections do not at all agree with the calculations of Bienstock *et al.* [4]. Their predicted second local maximum of the cross section near 27 eV/amu is completely absent in our measurements.

At energies between 0.1 and 100 eV/amu, a comparison can be made with the fully quantal calculations of Herrero *et al.* [5]. Reasonable agreement between our results and theirs is found, especially if only the calculations for capture into the $N^+(^3D^o)$ state are considered, except for a theoretical peak at 0.25 eV/amu, which is not reproduced experimentally, as will be discussed below. Inclusion of the $N^+(^3P^o)$ channel increases the electron-capture cross section above 10 eV/amu, and makes the agreement poorer. It should be noted, though, that Herrero *et al.* [5] did not opti-

mize their calculations at the high side of their energy range.

The strong upturn of the cross section at energies below 10 eV/amu could be an indication of the increasing importance of trajectory effects caused by the ion-induced dipole interaction. Such trajectory effects have very recently been identified for the $Si^{4+}+D$ system [2]. They not only lead to an increase of the cross section (scaling as $1/v$ as $v \rightarrow 0$), but also to a kinematic isotope effect [2]. Since at the lowest energies our experimental data are for D, and theory is for H solely, a comparison between theory and experiment should be treated with care. If trajectory effects do play a role, the electron-capture cross section for collisions involving D will be lower than the one involving collisions with H. From this point of view, it is regrettable that no theoretical data are available at low energies for $N^{2+}+D$. Since for isotope D no peak is observed, theory might help explain the origin of the peak at 0.25 eV/amu predicted by Herrero *et al.* [5], namely whether it really exists (it should then be isotope dependent), or whether it is a theoretical artifact (for example, resulting from not taking into account enough partial waves at very low energies). At present, our experiment cannot give the answer, because of the limited dynamic range of the ECR source, which means that the low energy region for collisions of N^{2+} ions with H cannot be reached.

Finally, we point out that Fig. 2 suggests that the calculations of Bienstock *et al.* [4] might follow the aforementioned $1/v$ scaling. However, their data show yet another local maximum in the cross section at 10 meV/amu. This is below the ultimate low-energy limit of our present setup, which is approximately 20 meV/amu [2].

IV. CONCLUSIONS

In the collision energy range of 0.1–4500 eV/amu, independent, absolute, total electron-capture cross sections for $N^{2+}+H(D)$ have been determined experimentally, using a merged-beams setup. Above 2000 eV/amu excellent agreement is found with previous experimental data of Phaneuf *et al.* [8] and Wilkie *et al.* [6]. Surprisingly, our results are about 30% below the measurements of Wilkie *et al.* [6] and Seim *et al.* [9] between 200 and 2000 eV/amu. Such a disagreement could be ascribed to the different experimental techniques used. We suggest that a (small but) nonnegligible fraction of vibrationally excited hydrogen molecules in the two latter experiments could be responsible for a significant contribution to the measured cross section. Unfortunately, to the best of our knowledge, no data exist for the $N^{2+}+H_2$ ($v>0$) collision system to check whether its electron-capture cross section indeed shows a maximum in the 200–2000 eV/amu energy range. If correct, we have shown that for some systems, especially when the cross section for electron capture from atomic hydrogen is low, there may be regions of the collision energy where hydrogen furnace measurements overestimate the cross section.

In addition, no evidence has been found for any clear maxima or oscillatory structure in the cross section below 100 eV/amu, in contrast with theoretical predictions of Bienstock *et al.* [4] and Herrero *et al.* [5]. Apart from this, it must be said that the results of the low-energy calculations of Herrero *et al.* [5] for the $N^+(^3D^o)$ channel agree better with our present measurements than those of Bienstock *et al.* [4] in the energy range where the calculations overlap.

As was mentioned in the introduction, the measurement of low-energy electron-capture cross sections can be used as a benchmark for theoretical models. This certainly holds for $N^{2+}+H(D)$: accurate theoretical calculations that cover a wide energy range of, say, 0.1–10000 eV/amu are still needed to resolve the puzzling discrepancies observed in this paper.

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