Low-energy electron capture by N^{4+} ions from H atoms: Experimental study using merged beams and theoretical analysis by molecular representation

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Measurements and theoretical analyses of total electron-capture cross sections for collisions of N^{4+} with ground-state hydrogen (deuterium) are reported in the energy range 1–300 eV/u. The present measurements have reduced relative uncertainty compared to previous absolute measurements at Oak Ridge National Laboratory [Huq *et al.*, Phys. Rev. A **40**, 1811 (1989)] and are used for detailed comparison with more recent coupled-channel molecular-orbital calculations [Shimakura *et al.*, Phys. Rev. A **45**, 267 (1992); Zygelman *et al.*, *ibid.* **46**, 3846 (1992)]. The most striking difference between the calculations was the increasing trend in the cross section for collision energies below 4 eV/u, as estimated by Zygelman *et al.* using only singlet states of the quasimolecule and the decreasing trend predicted by Shimakura *et al.* using both singlet and triplet states. The latter decreasing trend is consistent with the previous measurement. At 0.5 eV/u, the predicted singlet cross sections differ nearly by a factor of 2. Possible origins for this difference are explored. Strong structure is predicted in the cross section of Zygelman *et al.* and, to a lesser extent, in that of Shimakura *et al.*; however, the phase of the structure is different in the two theories. Structures observed in the measurement at the higher collision energies are not in complete harmony with either prediction.

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I. INTRODUCTION

The capture of electrons from neutral atoms by low-energy multicharged ions continues to receive considerable attention. The current interest in such processes arises from their practical applications in studies of controlled thermonuclear fusion, research on x-ray lasers, radiation research, astrophysics, and development of multiply charged ion sources. For partially stripped, multiply charged ion–hydrogen-atom systems, a number of groups have carried out theoretical calculations of various degrees of sophistication. In particular, the collision system

$$N^{4+}+H\ (D)\rightarrow N^{3+}+H^{+}\ (D^{+}) \tag{1}$$

has been the subject of recent theoretical efforts partly because of the existence of previous low-energy total-electron-capture measurements. The N^{4+} ion has a Li-like electronic structure, and no long-lived metastable states are known to exist to complicate comparison with theory. Previous absolute measurements [1] were performed with the Oak Ridge National Laboratory (ORNL) ion-atom merged-beam apparatus [2] in the energy range between 1 and 1000 eV/u using both H and D neutral beams. No other low-energy measurements are known that extend below 1 keV/u.

Heretofore, three different theoretical studies can be compared with each other and the experimental data. In the calculation of Fiekert *et al.* [3], which was based on a molecular-expansion method and was limited to energies below 2.5 eV/u, the molecular electronic structure was determined by using the *ab initio* configuration-interaction (CI) method with a Gaussian basis set. Only configurations that correspond to states with one active electron were considered. An approximate estimate of the coupling matrix elements was made from these potential energies. The quantum-mechanical, close-coupling calculation performed included four states that asymptotically converge to the initial state and three final states denoting capture to the $2s3s$, $2s3p$, and $2s3d$ configurations. These calculations overlap with the experimental data only at the lowest energies; they predict that the cross section will increase sharply below 1 eV/u.

A recent calculation by Shimakura, Itoh, and Kimura [4] in the collision-energy range between 1 eV/u and 10 keV/u used semiclassical and quantum-mechanical molecular-orbital (MO) methods with atomic-type electron translation factors (ETF's). The calculations included 12 channels and three channels for the semiclassical and quantum-mechanical approaches, respectively, for both singlet and triplet manifolds. The semiclassical results above 20 eV/u exhibited structures in the total cross section. These structures were attributed to an interference effect observed mostly in capture to triplet states. Full quantal calculations below 20 eV/u showed a decreasing cross section for both triplet and singlet cases, in

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agreement with the previous experimental data for N^{4+} [1].

Another recent calculation carried out by Zygelman *et al.* [5] for collision energies between 0.01 and 600 eV/u was based on a molecular-state expansion method using only singlet states without ETF's. A spin-coupled valence-bond CI method was used to construct *ab initio* potential curves and, hence, nonadiabatic-coupling matrix elements. The calculated cross sections exhibited large variations with energy, the physical interpretation of which was a manifestation of Stueckelberg oscillations. However, the experimental data were inadequate to resolve the predicted structure. Agreement with the previous measurements [1] was good between 4 and 180 eV/u where singlet and triplet cross sections are not expected [5] to be sensitive to small differences in corresponding adiabatic potential. The previous experimental data had a decreasing cross section with decreasing collision energy, in contrast to this calculation's prediction of a generally constant cross section below 20 eV/u with an increasing trend below 4 eV/u.

To investigate the origin of this discrepancy between the two sets of calculations and the previous experimental data, we undertook a combined experimental and theoretical study of capture of electrons in low-energy collisions of N^{4+} ions with ground-state H (D) atoms. We report here our findings. The present apparatus has increased sensitivity, resulting in more data points with lower relative uncertainties and an increased angular acceptance to ensure collection of signal at the lower energies. Both the previous and present measurements were performed with D rather than H to maximize signal collection. Measurements reported here are relative, because of the use of an uncalibrated multichannel plate detector, but were put on an absolute scale by normalization to the previous ORNL absolute measurements. The present data are sufficient both to observe the predicted oscillations if they exist and to better establish the trend of the total cross section at lower energies.

Additional fully quantal coupled-channel MO calculations were performed at the lower energies. The essential approximations used in the theory were the same as in our previous calculation [4], except that we enlarged the MO basis size to ensure the convergence of the cross section. We also compared various aspects of the approximations and numerical details used in the two calculations in a step-by-step manner.

II. EXPERIMENTAL METHOD

A. Merged-beam technique

Total electron-capture cross sections were measured at the low energies by using the merged-beam method. Only a brief description is presented here. For more details on the technique, the reader is directed to [2] and to previous merged-beam measurements [1] on the N^{4+} system. In this technique, fast (keV) beams of neutral atoms and multicharged ions are merged. The resulting relative velocities of the two beams can be "tuned" over a very large range. The collision energy $E_{\rm rel}$ in eV/u corresponds to the relative interaction energy of the two beams divided by the reduced mass and is given by

$$E_{\rm rel}=\left[\frac{E_1}{m_1}+\frac{E_2}{m_2}\right]-2\sqrt{[(E_1E_2)/(m_1m_2)]}\cos\theta\ , \quad (2)$$

where E_1 and m_1 correspond to the energy (eV) and mass (a.u.) of the neutral beam, and E_2 and m_2 to those of the multicharged ion beam. The angle θ is the merge angle of the two beams. To first order, θ is equal to zero. Figure 1 is a simplified diagram of the apparatus. The multicharged ion beam of N^{4+} produced by the ORNL CAPRICE electron cyclotron resonance (ECR) ion source is merged electrostatically with a neutral D beam. The merged beams interact in a field-free region for a distance of 47 cm, after which the primary beams are magnetically separated from each other and from the product or "signal" D^+ ions. The N^{3+} product of the reaction is not measured separately but is collected with the primary N^{4+} product in a large Faraday cup. The neutral-beam intensity is measured by secondary-electron emission from a stainless-steel plate, and the signal D^+ ions are recorded by a circular multichannel plate detector with a 4-cm active diameter. A 99.98% pure ground-state beam of D atoms is produced by passing an 8-keV beam of D^- ions through the optical cavity of a 1.06-μm Nd:YAG (yttrium aluminum garnet) laser, where up to 600 W of continuous power circulates and typically 0.5% of the negative ions undergo photodetachment. An electric-field ionizer is used to quench excited D atoms whose electrons are in high-n shells and which are produced by collisional stripping of D^- on background gas. A nearly parallel beam of D atoms is produced having a diameter of 2–4 mm full width at half maximum (FWHM) and an equivalent intensity of 10–20 (particle) na. The divergence of this beam is typically less than 0.2°. The 55- to 85-keV, 2- to 5-μA beam of N^{4+} ions produced by the ORNL ECR ion source has a typical diameter of 6–8 mm FWHM in the merge path and a divergence of less than 0.5°. The finite divergence of the primary beams results in a distribution of merging angles, creating a small absolute shift and energy spread in the collision energy. Electron-capture cross sections are determined by

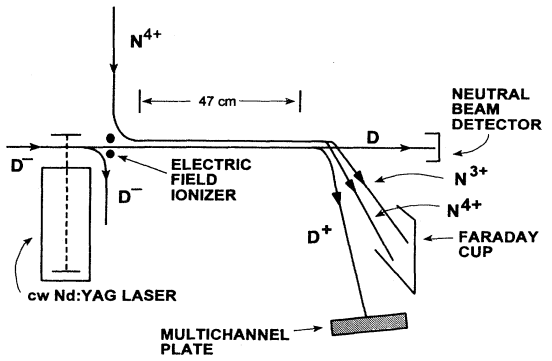

FIG. 1. Schematic diagram of the ion-atom merged-beam apparatus for the $N^{4+}+D$ collision system.

measuring the rate of D^+-ion production by the beam-beam interaction over the merge path. The cross-section value σ is determined at each velocity from directly measurable parameters by the following formula:

$$\sigma = Rqe^2\gamma v_1 v_2 / [I_1 I_2 \varepsilon \langle F \rangle L v_r] . \quad (3)$$

Here R is the signal count rate; q is the charge of the ion; e is the electronic charge; γ is the secondary-electron emission coefficient of the neutral-beam detector; I_1 and I_2 are the currents of the two beams; ε is the efficiency for detecting the product D^+; $\langle F \rangle$ is the average form factor, which is a measure of the overlap of the beams over the merge path of length L; v_1 and v_2 are the velocities of the beams; and v_r is the relative velocity between beams. The integrated three-dimensional form factor is estimated from two-dimensional measurements of the overlap at three different positions along the merge path. γ was measured *in situ* [2] and found to be 1.15 ± 0.06 for these measurements with a D beam of 8 keV.

The D^+ product ions were recorded by monitoring the strobe output from a two-dimensional detector system, Quantar Technology model 3394A. The detector consisted of two multichannel plates (with the front of the first plate operating at ground potential) and a resistive anode with a 4-cm active area. The strobe output corresponded to a busy signal for the position computer and did not include position information. Two-dimensional information was used during tuning of the background on the detector but not during data acquisition. The total counting efficiency (electronics plus detector efficiency) was determined by normalizing these data to the previous absolute measurements. This efficiency ε was determined to be 0.68, which is slightly greater than the open area (0.63) of the multichannel plates. The signal rate R was measured from background by using a two-beam-modulation technique. Backgrounds on the order of 10 kHz were produced by the fast-neutral-beam stripping of D on the background gas in the merged path, where pressures were on the order of 1.3×10^{-8} Pa. Photon backgrounds on the order of 80 Hz resulted from collection of N^{4+} in the Faraday cup. Signal rates on the order of 30 Hz were observed at the higher collision energies, with the signal decreasing to a few Hz at the lower energies due to both a drop in the cross section and the decrease in the number of collisions in the merge path.

B. Beam purity of D

As was previously observed [6] in merged-beams studies of $[O^{5+} + D]$ at collision energies below 10 eV/u, a very small (0.02%) Rydberg population in the D beam can produce a significant beam-beam signal compared to that due to capture from the ground state. The excited states of D are created by collisional stripping of the D^- beam on the background gas. Some of the excited D atoms which have electrons in n shells between 11 and 24 have trajectories that allow them to merge with the multicharged ions and, if electron loss occurs, to produce a beam-beam D^+ signal. The cross section for electron removal from an excited state of D is typically greater by a factor of 10^5 than the capture cross section from the ground state. These contributions from excited states, which can be a significant fraction of the beam-beam signal, were studied in some detail by using this apparatus [7]. To correct for the signal due to the excited states, the beam-beam signal was measured with the laser on and then off, so that the appropriate difference between the signals corresponded to the signal due to the ground state. To reduce the Rydberg population, the D beam was made to pass through a field ionizer (see Fig. 1) that ionized the higher-n levels of the excited D, causing them to be swept out of the neutral beam. The electric field required to field-ionize a particular n shell in D is approximated by the following semiempirical relation (see Ref. [7]):

$$E(\text{kV/cm}) = [6.25 \times 10^5 \text{ (kV/cm)}] / [n^4] . \quad (4)$$

For the measurements reported here, highly excited states of D were ionized down to $n = 12$ by using an applied electric field of 30 kV/cm. This reduction in the excited component of the D beam led to a beam-beam signal correction on the order of 5%.

C. Signal collection

Since the low-energy electron-capture collisions under study are exoergic and both products are positively charged, significant angular scattering of the D^+ can occur in the center-of-mass frame [8]. However, because of the kinematic frame transformation, this angular scattering is significantly compressed in the laboratory frame, the frame in which the products are collected. The angular acceptance of the apparatus in the laboratory frame is currently 2.3°, as determined by ray tracings and verified by comparison of data to angular-scattering calculations for the O^{5+} system (see Refs. [6,9]). From this estimate, one can determine the maximum angle into which the product D^+ can be emitted in the center-of-mass frame and still be collected. The final velocity of the D^+ product is taken to be the sum of the initial velocity and the velocity "kick" provided by the exoergicity of the reaction. Due to the conservation of linear momentum, this velocity "kick" to the detected collision product is reduced when D is used rather than H. Measurements, then, were performed with D to minimize the resultant angular scattering. For collisions with D, the angular acceptance in the center-of-mass frame is a function of collision energy, the center-of-mass velocity, and the exoergicity of the capture process. Figure 2 shows the angular acceptance in the center-of-mass frame in the forward direction as a function of collision energy for capture into the $N^{3+}(1s^2 2s 3d)\ ^1D$ configuration, which has an exoergicity of 10.3 eV. For scattering in the backward direction the angular acceptance is similar, because only the velocity component perpendicular to the beam direction leads to a loss of signal. The figures was constructed for an 8.0-keV D beam, the neutral beam used in the measurements. Figure 2 shows that the angular acceptance improves with decreasing collision energy and for this capture channel below 10 eV/u the angular acceptance approaches 90°; hence all product D^+ ions are collected. Also plotted is the angular acceptance of the previous ap-

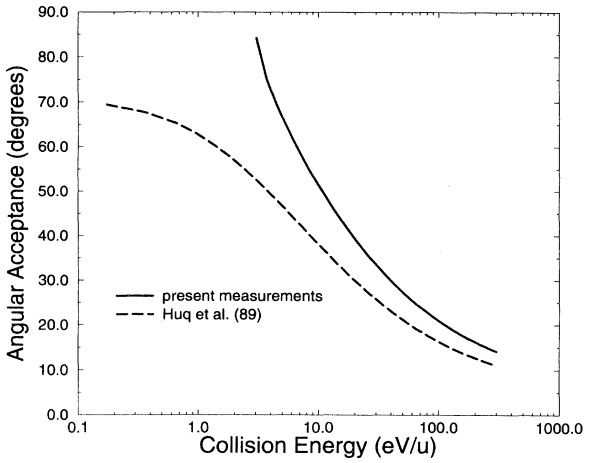

FIG. 2. Angular acceptance in the forward direction in the center-of-mass frame as a function of collision energy for measurements of the $N^{4+}+D$ system, with an 8.0-kV D beam ($Q=10.3$ eV). Angular acceptance in the backward direction is similar but is not shown.

paratus, which had an estimated angular acceptance in the laboratory frame of 1.8°. One can see that the angular acceptance has improved significantly, especially below 10 eV/u. Unfortunately, detailed angular-scattering calculations are not available for this collision system, so no guarantee can be made that all the signal is collected. However, the present angular acceptance was found sufficient [6,9] for the angular scattering in the $[O^{5+}+D]$ collision system.

D. Uncertainties

The voltage dividers that measure the accelerating potentials of the two sources are calibrated to each other within a relative accuracy of 2 V. The absolute voltage is determined within 0.05%. These accelerating potentials are modified by the plasma space potential of both the ECR and duo-plasmatron ion sources. Estimates [10] are $+20\pm10$ V per charge and $+10$ V, respectively. Both of these plasma potentials can be used in Eq. (2) to estimate the absolute shift in energy. As Eq. (2) shows, there is also a shift in collision energy that is due to nonzero merging angles. The bulk of collisions are estimated to occur with a merge angle of $0.35°\pm0.35°$. For energies 1 eV/u and greater, the resultant total absolute shift in collision energy, which is the sum of all the contributions, is of little significance (on the order of 1% of the collision energy). There is also a spread in collision energy due to the spread in merging angle. Like the shifts in energy, this spread is of significance only below 1 eV/u. For the measurements reported here, this correction is also on the order of 1% of the collision energy. Further details can be found elsewhere [10].

Relative uncertainties in the measured signal are in large part due to the statistical uncertainties in separating the beam-beam signal from the backgrounds. Occasionally spatial instabilities in the beams that are beyond our ability to monitor will add an additional component (in quadrature) due to changes in the beam-beam overlap. A detailed discussion of these uncertainties can be found in Ref. [2]. These measurements were placed on an absolute scale by normalizing to our previous absolute measurements.

III. THEORETICAL MODELS

First, we briefly outline our model used in the calculation, with particular emphasis on its approximations. Secondly, we discuss the various differences between the present results and those of Zygelman *et al.* [5] that are attributable to features of the two models. Our theoretical model and that of Zygelman *et al.* are essentially the same and are based on a molecular-orbital expansion method within a fully quantal representation. The procedure consists of two major tasks, namely, (i) determination of molecular states by using an *ab initio* CI procedure and (ii) determination of the scattering S matrix by solving quantal close-coupling equations. Note that our calculation and that of Zygelman *et al.* were carried out for the H target, whereas the D target was used experimentally. However, at a given collision velocity, as long as trajectory effects [6] due to the ion-induced dipole potential are not important, collisions with H or D should give the same value for the cross section. At 1 eV/u, which corresponds to the lowest measured collision energy, we confirmed this point numerically. The difference in the predicted cross section between collisions with H and D at 1 eV/u was found to be less than a few percent. Below 1 eV/u, though, trajectory effects may increase [6], which could result in a significant difference in the cross section, i.e., a smaller value for D and larger value for H. In addition, the difference between the binding energies of $H(1s)$ and $D(1s)$ is 0.0037 eV. For the collision-energy range considered here, this binding effect is not important.

A. Molecular states

The present procedure for determination of molecular states is a modified valence-bond CI method with a Gaussian-type pseudopotential representing the N^{5+} core and Slater-type orbitals (STO's) that are used as a basis set [4]. In contrast, the determination of molecular states by Zygelman *et al.* [5] is based on the spin-coupled valence-bond CI method with Gaussian-type orbitals as a basis set. Both use a reasonably large number of bases and CI's. Both sets of adiabatic potential curves for the initial $[N^{4+}+H]$ channel and the dominant charge-transferred $[N^{3+}(3d)+H^+]$ channel for the *singlet manifold* are illustrated in Fig. 3 in the vicinity of a strong avoided crossing near $R=8a_0$. A small difference is visible particularly in the region of $(7.5-8.5)a_0$, where the two curves approach. The energy splitting between the two potential curves at the avoided crossing as calculated by Zygelman *et al.* is somewhat narrower than that of our result. As they depart on both sides of the avoided crossing, the two sets of two curves are virtually identical

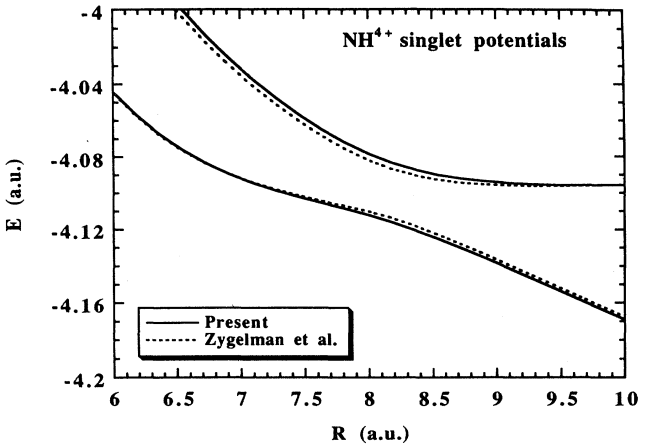

FIG. 3. Adiabatic potential curves for the initial [$N^{4+}+H$; upper curves] and capture [$N^{3+}(3d)+H^{+}$; lower curves] channels for the singlet manifold. Solid lines, present results; dashed lines, Zygelman *et al.* [5].

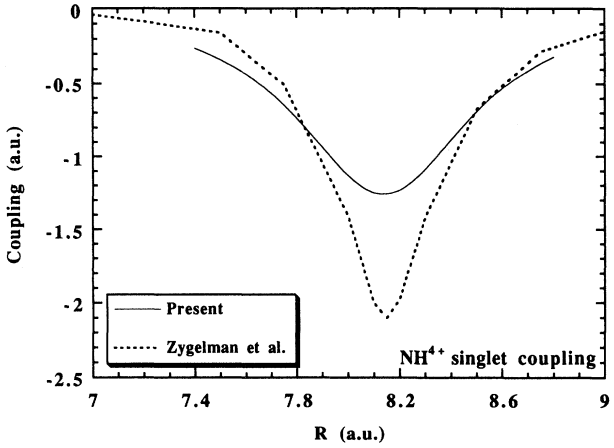

FIG. 4. Radial-coupling matrix elements between the initial and capture channels for the singlet manifold shown in Fig. 3. Solid line, present results; dashed line, Zygelman *et al.* [5].

and are not distinguishable graphically. Some representative energies from both calculations, along with those obtained from the asymptotic form of the polarization potential for the initial channel, are summarized in Table I. The initial-channel potential of Zygelman *et al.* is somewhat shallower than those of the other two sets. However, both calculations appear to reproduce reasonably well the asymptotic behavior of the polarization potential, which is essential for the correct description of low-energy collisions.

Radial couplings from two calculations for the singlet manifold are depicted in Fig. 4. These couplings were obtained numerically from the corresponding wave functions. Our peak-height value for the coupling is 1.27 a.u. at $R=8.14a_0$, which amounts to 60% of Zygelman's value. (That of Zygelman *et al.* is 2.10 a.u. at $R=8.15a_0$.) In our calculation, the coupling shown includes a correction for the ETF [11], which helped reduce the original peak height by about 20%. Zygelman *et al.* did not include the ETF effect. The agreement in energies for less dominant channels like $N^{3+}(3p)$ and $N^{3+}(3s)$ states is slightly better than that shown in Fig. 3. Consequently, the couplings that connect those channels were found to be in reasonable agreement in magnitude and shape, apart from the ETF effect. We also checked the quality of the molecular states we obtained (i.e., the values of the energies and couplings) by increasing the basis size to the $N^{3+}(n=2, n'=4$ and 5) levels, beyond the largest basis set used previously (52 STO's versus 38 STO's in our previous calculation). The location of the avoided crossing between the initial and the $N^{3+}(3d)$ channels was found to vary by less than 2% and the corresponding peak height in the coupling changes by about 3%.

TABLE I. Adiabatic potential values for the [$N^{4+}+H$] channel of the singlet manifold from the present work, that of Zygelman *et al.* [5], and the asymptotic form of the polarization potential. (Note that all values are normalized to that of the polarization potential at $R=30a_0$.)

	Energy (a.u.)		
R (a_0)	Present	Zygelman *et al.*	Polarization potential
10	0.003 608	0.003 581	
15	0.000 698	0.000 581	0.000 711
20	0.000 219	0.000 204	0.000 225
30	0.000 044	0.000 044	0.000 044

B. Collision dynamics

To solve the coupled equations, both calculations used the log-derivative method [12] after transformation of the coupled equations in an adiabatic representation to those in diabatic representation. In addition, we used the Numerov method [13] to check the numerical precision of solutions of the coupled equation obtained by the log-derivative method and found that both results from these two solvers agree within a few percent in the S matrix. We also solved the coupled equations by *using* Zygelman's tabulated potentials and coupling and reproduced their cross sections at 0.5, 1, and 5 eV/u. Our present calculation gives a slightly larger cross section than our previous results because we included additional channels and used a smaller mesh size. However, we essentially reproduced our previous results. The old calculation used a larger energy-mesh size and hence missed the oscillatory structures that are apparent in the new calculation. These test calculations appear to confirm that there were no numerical errors in the calculations of Zygelman *et al.* [5] and Shimakura, Itoh, and Kimura [4] in solving the coupled equations.

IV. RESULTS AND DISCUSSIONS

A. Theoretical results

On the basis of the various checks described above, we conclude that the difference seen in the two sets of cross sections may originate in the difference in the potential curves and the corresponding coupling matrix element, as described earlier. We extended our calculation below 1 eV/u and found that our preliminary cross section appears to decrease as energy decreases down to about 0.5 eV/u, when it gradually increases as the collision energy decreases further. Zygelman *et al.* [5] saw a similar trend, but it occurred at much higher energies, beginning as high as 4 eV/u. We also predicted similar oscillatory structures due to multichannel interferences (the Stuekelberg oscillation) throughout the energy region studied, although the oscillatory pattern and amplitude are significantly weaker in our calculation.

In the cross-section calculations, Zygelman *et al.* [5] included the singlet manifold *only*, while the present calculation considered *both* triplet and singlet manifolds. The present triplet, singlet, and total contributions separately are illustrated in Fig. 5 along with that of Zygelman *et al.* [5]. The present singlet cross section agrees reasonably well with that of Zygelman *et al.* above 3 eV. Below this energy, though, the significant difference in the two results begins to emerge. At 0.5 eV/u, the results differ by more than a factor of 2. Note that the triplet contribution was found to have only a slightly different energy dependence than the singlet manifold. Beginning with roughly 70% of the total cross section at 10 eV/u, the triplet contribution decreases as energy decreases, falling to about 60% at 1 eV/u. The neglect of the triplet manifold in the calculation by Zygelman *et al.*, however, did not contribute to the present discrepancy, because in the present calculation, both triplet and singlet cross sections were found to decrease. Nevertheless, the triplet manifold should be treated equally for an accurate description of collision dynamics. In Table II, we summarize the primary differences in approximations and procedures used in the two theoretical approaches.

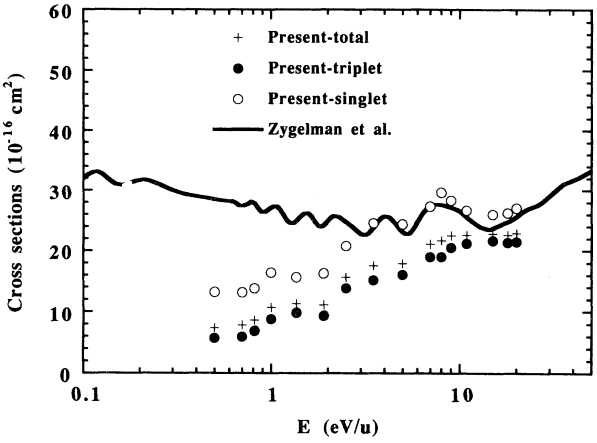

FIG. 5. Theoretical capture cross sections below 20 eV/u. Present results: solid circle, triplet contribution; open circle, singlet contribution; and cross, total cross section, i.e., [$\frac{3}{4}$ triplet + $\frac{1}{4}$singlet]. Solid line, Zygelman *et al.* [5] (singlet only).

B. Experimental results

The measured total electron-capture cross sections for $N^{4+}+D$ are presented in Table III. The total uncertain-

TABLE II. Summary of approximations used in the two theoretical approaches.

	Present work	Zygelman *et al.* [5]
Molecular state		
(i) Method	Modified valence-bond CI method	Spin-coupled valence-bond CI method
(ii) Pseudopotential	N^{5+} core	None
(iii) Basis	Slater-type orbital	Gaussian-type orbital
(iv) Position and height of dominant coupling	$8.14a_0$ 1.27 a.u.	$8.15a_0$ 2.1 a.u.
(v) States included	$N^{4+}+H$ initial $N^{3+}(3d,3p,3s)+H^+$	$N^{4+}+H$ initial $N^{3+}(3d,3p,3s)+H^+$
(vi) Triplet and singlet manifold	Both triplet and singlet manifolds	Singlet manifold only
Dynamics		
(i) Method	Fully quantum-mechanical close-coupling method	Fully quantum-mechanical close-coupling method
(ii) ETF	Atomic-type ETF	None
(iii) Numerical method	Log-derivative method for diabatic representation	Log-derivative method for diabatic representation

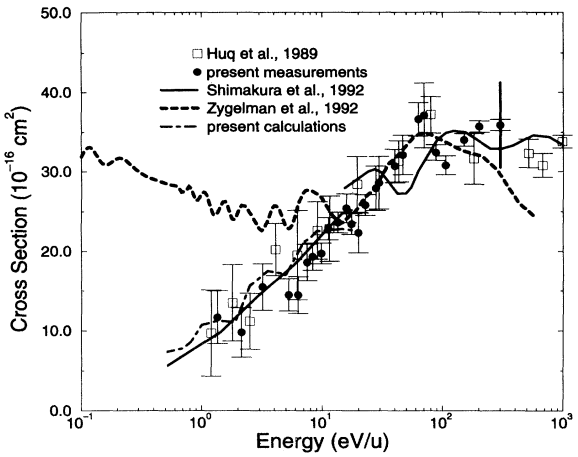

FIG. 6. Present results compared with previous measurements and calculations for $N^{4+}+D$ collisions. All vertical error bars denote relative uncertainties estimated at a 90% confidence level. At 301 eV/u an additional vertical error bar denotes the absolute uncertainty at a 90% confidence level. Experimental results: solid circle, present work; open square, Huq *et al.* [1]. Theoretical results; solid line, Shimakura, Itoh, and Kimura [4]; dashed line, Zygelman *et al.* [5]; and dashed-dotted line, present work.

ties correspond to a quadrature sum of the relative and absolute uncertainties estimated at the 90% confidence level. These measurements were put on an absolute scale by normalization to the previous absolute measurements at 43 eV/u. The normalization procedure resulted in an absolute uncertainty of 15%. The present measured data, the previous absolute measurements by Huq, Havener, and Phaneuf [1], our present theoretical results, the previous calculations of Shimakura, Itoh, and Kimura [4], and the calculations of Zygelman *et al.* [5] are summarized in Fig. 6. The present measurements are plotted with vertical error bars that denote the relative uncertainties estimated at the 90% confidence level, as presented in Table III. At a collision energy of 301 eV/u, an additional vertical error bar denotes the total uncertainty of the measurements. As can be seen in the figure, the energy dependence of the present measurements agrees very well with the previous ORNL data.

The data indicate that the increased angular acceptance of the apparatus and the reduced D excited-state contribution did not affect the previously observed decreasing trend of the cross section with decreasing collision energy. This trend was predicted by both the present calculation and the previous one of Shimakura, Itoh, and Kimura [4], in contrast to the estimate of Zygelman *et al.* [5]. According to our present theoretical study, the origin of this difference may be attributed to the difference in the adiabatic potential curves and the coupling between the initial and the dominant $N^{3+}(3d)$ channels, as exemplified in Figs. 3 and 4. The weaker oscillations in our present calculations, as compared to those of Zygelman *et al.*, may be a washout effect due to the inclusion of the triplet and singlet manifolds. Weak oscillatory structures are suggested in the present mea-

TABLE III. Measurements of total electron-capture cross sections for $N^{4+}+D$. All uncertainties are estimated at the 90% confidence level. The total uncertainty represents the quadrature sum of the relative and absolute uncertainties.

Collision energy (eV/u)	Cross section (10^{-16} cm^2)	Relative uncertainty (10^{-16} cm^2)	Total uncertainty (10^{-16} cm^2)
1.35	11.7	3.3	3.7
2.13	9.8	3.1	3.4
3.19	15.5	2.9	3.7
5.25	14.5	2.0	3.0
6.29	14.5	2.3	3.2
7.45	18.6	2.3	3.6
8.30	19.3	1.7	3.4
9.81	19.7	1.3	3.2
11.3	22.8	1.4	3.7
13.5	23.6	1.3	3.8
15.8	25.4	1.8	4.2
17.5	23.4	1.3	3.7
19.9	22.3	2.5	4.2
22.7	25.8	1.3	4.1
27.7	27.9	2.8	5.0
29.4	28.6	3.3	5.4
40.0	30.7	2.1	5.1
46.6	32.1	2.5	5.4
63.1	36.6	2.1	5.9
70.2	37.1	4.1	6.9
88.1	32.4	1.0	5.0
106	30.8	1.2	4.8
151	34.0	0.8	5.2
201	35.7	0.7	5.4
301	35.9	0.7	5.4

surements below 20 eV/u, but they are of little significance when compared to the estimate of relative uncertainty.

The maximum in the cross section occurred at 70 eV/u, in good agreement with the calculations of Zygelman *et al.* [5]. However, these calculations predicted that the cross section sharply drops toward higher energies, in contrast to the ORNL merged-beams measurements or the measurements at higher energies of Crandall, Phaneuf, and Meyer [14] and Seim *et al.* [15]. At energies above 500 eV/u, this discrepancy is attributed [5] to the role of additional channels. The calculations of Shimakura, Itoh, and Kimura [4] are in good accord with the energy dependence of these high-energy measurements. They predict that the cross section will show strong oscillations at energies above 20 eV/u but remain essentially constant toward the higher energies. The structure, though, when compared to the present measurements, is slightly out of phase. Because many channels are coupled simultaneously in this energy domain, the oscillatory structure is sensitive to the details of the potentials and couplings included.

V. CONCLUSIONS

We have carefully examined electron capture by N^{4+} ions with H (D) atoms both experimentally and theoretically, in order to identify the origin of the difference between two theoretical results, those of Shimakura, Itoh, and Kimura [4] and Zygelman *et al.* [5]. The present experiment reproduced the previous ORNL data, and the present theory reproduced the previously reported results of Shimakura, Itoh, and Kimura. The difference in the two predicted trends in the singlet cross section toward lower energies appears to be due to a small difference in the molecular states obtained by the two different CI approaches. In addition, the omission of the ETF's in one calculation increases to some extent this difference between the calculations. Also, the strong structure predicted by the calculations of Zygelman *et al.* below 10 eV/u is not observed. At higher energies, the maximum in the cross section shows good agreement with the calculation of Zygelman *et al.*, while the general trend of the data is predicted by the calculations of Shimakura, Itoh, and Kimura. Predicted oscillation patterns are slightly out of phase with that observed.

The discrepancy between two essentially similar methods, and the somewhat smaller discrepancy at higher collision energies, emphasize that fully quantal, coupled-channel calculations are by no means routine to perform and that continued comparison with experiment is necessary for a correct understanding of collision dynamics. The recent improvements in the ion-atom merged-beam apparatus reduced the experimental uncertainties, allowing a critical comparison with predicted total capture cross sections. Such comparison was previously possible only with state-selective measurements, which, at these energies, are not readily available.

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