Electron Capture by Ne^{4+} Ions from Atomic Hydrogen

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Abstract

Using the Oak Ridge National Laboratory ion-atom merged-beams apparatus, the absolute total electron-capture cross section has been measured for collisions of Ne^{4+} with hydrogen and deuterium at center-of-mass energies between 0.10 and 1006 eV/u. Comparison to previous measurements shows large discrepancies between 80 and 600 eV/u. For energies below ∼1 eV/u, a sharply increasing cross section is attributed to the ion-induced dipole attraction between the reactants. Multichannel Landau-Zener calculations are performed between 0.01 and 5000 eV/u and compare well to the measured total cross sections. Below ∼5 eV/u, the present total cross section calculations show a significant target isotope effect. At 0.01 eV/u, the H/D total cross section ratio is predicted to be ∼1.30 where capture is dominated by transitions into the Ne^{3+} $(2s^2 2p^2 3d)$ configuration.

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I. INTRODUCTION

Studies of electron-capture (EC) processes are motivated by the fact that they constitute reaction channels of fundamental importance in plasma environments. In astrophysics, EC by multi-charged ions from atomic hydrogen are important in planetary nebulae with hot central stars. In that environment the large number of stellar photons with energies well above the H ionization limit (where the photoionization cross section is low) results in a significant fraction of H in the nebula being neutral [1]. Temperatures in planetary nebulae are typically on the order of 10,000-30,000 K. Ne emission lines are used to measure temperature and density and to infer the elemental abundances in the nebula (see, e.g., [2]). In fusion energy research, EC cross sections for energies between ∼1 to ∼100 eV/u are necessary for accurate modeling and diagnostics of the edge plasma. Injection of Ne into the DIII-D device, the third generation tokamak developed by General Atomics in San Diego, CA, is used to enhance the radiation in the core plasma reducing the heat flux to the plasma facing surfaces. Impurity injection thus improves the energy confinement and overall plasma performance. Accurate modeling of the Ne ion charge balance and radiative cooling in these plasmas requires reliable EC cross section data for a wide range of energies and Ne ionization stages [3]. Systematic studies of the low energy EC for Ne^{2+} + H and Ne^{3+} + H have recently been reported [4, 5]

We are unaware of any published theoretical studies for low-energy electron capture in the Ne^{4+} + H collision system. Previous cross section measurements performed by Can *et al.* [6], Huber [7] and Seim *et al.* [8] at relatively high energies (i.e., above 60 eV/u) over a limited energy range indicate a decreasing cross section toward lower energies. At lower collision energies (eV/u), the ion-induced dipole interaction between reactants is important. The attractive force due to this potential, and the resulting acceleration of the particles toward each other, significantly modify the reactant trajectories. This results in the incident trajectories accessing internuclear distances smaller than the initial impact parameter. At low enough energies, these trajectory effects can dominate the electron transfer process and lead to enhanced cross sections [9]. Several systems have been investigated using the ORNL ion-atom merged-beams apparatus which show such enhancements [10]. Ion-induced dipole enhancements also lead to significant target isotope effects [11, 12].

Here, using the ion-atom merged-beams apparatus, the absolute total cross section is measured for the electron transfer process

$$Ne^{4+} + H \rightarrow Ne^{3+} + H^{+} \tag{1}$$

over four decades of collision energy, from 0.10 to 1006 eV/u. Below 250 eV/u, D is used instead of H for experimental convenience. For comparison, Multi-Channel Landau-Zener (MCLZ) calculations for collisions with hydrogen and deuterium are performed over the energy range of 0.01 to 5000 eV/u. Due to the good agreement between the measured and calculated total cross sections, MCLZ calculations are also used to predict state-selective cross sections.

II. EXPERIMENTAL APPROACH

In the merged-beam technique, relatively fast (keV) beams are collinearly merged producing a large dynamic range of collision energies in the center-of-mass (c.m.) [13]. In the present investigation, a Ne^{4+} beam with energies of 52-88 keV is merged with D beams at energies of 6.95 - 9.55 keV and an H beam at an energy of 6.70 keV, thereby allowing c.m. collision energies in the range of 0.10-1006 eV/u. The apparatus is depicted schematically in Fig. 1 and has previously been described in detail [10, 13].

As shown in Fig. 1, a neutral ground state H atom beam is obtained by photodetachment of a H^- beam as it crosses the optical cavity of a 1.06 μm cw Nd:YAG laser where kilowatts of continuous power circulate. The H^- beam is extracted from a duoplasmatron source. Collisional detachment of the H^- beam on background gas results in a small fraction (0.01%) of excited states in the H beam. The H beam obtained is nearly parallel (the divergence being less than 0.15°) with a beam diameter of 2 mm and intensities ranging from 10-20 nA. Deuterium is used instead of hydrogen for c.m. energies below 250 eV/u to maximize the angular acceptance of the $H^+(D^+)$ detector [13, 14].

The Ne^{4+} beam is produced by the ORNL CAPRICE ECR ion source [15] with an intensity of approximately 4 μA, a diameter of 2-4 mm (FWHM), and a divergence less than 0.25°. The purity of the Ne^{4+} beam from the ORNL CAPRICE with respect to metastable states was previously explored by Bannister [16] using electron-impact ionization. A small cross section observed below the expected ground state threshold suggests that only a few percent of the beam is in the metastable state but the statistical uncertainty of the signal in the electron-impact ionization measurements make a reasonable estimate of the metastable fraction impractical.

The Ne^{4+} beam is electrostatically merged with the neutral H beam (see Fig. 1). The ion and atom beams interact along a field-free region of 47 cm, after which the H^+ product ions are magnetically separated from the primary beams. The merge path is maintained under ultra-high vacuum conditions to minimize the H^+ background generated by stripping of the H beam as it travels through the background gas. To maximize the angular acceptance of H^+ by the H^+ detector, a cylindrical Einzel lens is placed near the end of the merge path. Due to limited magnetic dispersion, the heavy product of the reaction, Ne^{3+}, is not measured but is collected along with the primary ion beam, Ne^{4+}, in a large Faraday cup. Since only the H^+ signal is measured, the apparatus actually measures electron loss, the sum of electron capture and ionization. However, ionization at these energies is negligible compared to electron capture [17]. The H neutral beam is monitored by measuring secondary emission from a stainless steel plate. The product signal (H^+) ions are detected by a channel electron multiplier. The signal rate (Hz) is extracted from the background (kHz) by a two-beam modulation technique [13]. To correct the signal rate for the small fraction of excited H, the signal is measured with and without the laser on. The difference between the signals corresponds to the signal due to the H ground state collisions.

The absolute electron-capture cross section is determined at each velocity from directly measurable parameters with the formula

$$\sigma = \frac{R\gamma q e^2 v_1 v_2}{I_1 I_2 v_r \langle F \rangle} \tag{2}$$

where R is the signal count rate, γ the secondary electron emission of the neutral detector, q the charge of the ion, e the electron charge, v_1 and v_2 the velocities of the beams, I_1 and I_2 the intensities of the two beams, v_r the relative velocity between beams, and $\langle F \rangle$ the average form factor, which is a measure of the overlap of the two beams. The form factor is determined from two-dimensional measurements of the overlap of the two beams at three different positions along the merge path. The secondary electron emission coefficient, γ, is measured *in situ* as described previously [13]. The velocities v_1 and v_2 are calculated from the energies of the beams, which include the estimated plasma potential shifts of the two sources. The relative velocity, v_r, is calculated from the velocities of the beams and the measured merge angle between the beams for energies less than 1 eV/u (see, e.g., Ref. [11]).

III. MULTICHANNEL LANDAU-ZENER CALCULATIONS

For the low-energy collisions considered here, molecular-orbital close-coupling (MOCC) calculations are considered to be the most appropriate and accurate theoretical approach. However, the necessary molecular potentials were not available for this system, so the simplier and more schematic multi-channel Landau-Zener (MCLZ) approach was used. While MCLZ calculations are not always reliable, in systems where the crossings as a function of internuclear separation R between initial and final states of the electronic potentials are localized and are dominated by radial couplings, the model can provide a good estimate of electron capture. Specifically for such systems where the MCLZ predictions are borne out by experiment, MCLZ calculations can be used to extend the cross sections to lower energies unaccessible by experiment and to explore isotope effects.

Here, MCLZ calculations were performed following the prescription of Butler and Dalgarno [18], including the H static dipole polarizability in the radial velocity relation, and the multichannel probability from the formulation of Janev *et al.* [19]. Empirical potentials and couplings were determined following Butler and Dalgarno. States of the separated atoms in the initial channel, $Ne^{4+}(2s^2 2p^2\ ^3P)$ + H(1s ^2S), correlate to the four molecular states $^2\Sigma^-$, $^2\Pi$, $^4\Sigma^-$, and $^4\Pi$ with approach probability factors of 1/9, 2/9, 2/9, and 4/9, respectively. For the electron capture channels, all LS terms were included for Ne^{3+} + H^+ which had avoided-crossing distances between 3 and 18 a_0 which included Ne^{3+} single-excitation configurations of $2s^2 2p^2 3s$, $2s^2 2p^2 3p$, $2s^2 2p^2 3d$, and $2s^2 2p^2 4s$ and the multiple-excitation terms $2p^5\ ^2P^o$ and $2s2p^3 3s\ ^4S^o$. Since rotational couplings were neglected, this resulted in four spin-symmetry MCLZ calculations, one for each of the molecular states above, with 8, 14, 6, and 5 electron capture channels, respectively. The MCLZ calculations were repeated for the D target.

IV. RESULTS AND DISCUSSION

Table I lists the measured absolute total EC cross section for Ne^{4+} + H as a function of collision energy. The statistical and total uncertainties are estimated at the 90% confidence level. The total uncertainty corresponds to a quadrature sum of the statistical and systematic errors. For energies less than 1 eV/u, the uncertainty in c.m. collision energy due to the energy spread of the D and Ne^{4+} primary beam and the spread in the merge angle (see Ref. [11]) is also shown in the first column of Table I. Fig. 2 compares the measured cross sections to the present MCLZ calculations and to previous experiments. While Fig. 2 shows the present measurements to be in agreement with the results of Seim *et al.* [8], there is significant discrepancy with the measurements of Can *et al.* [6] and Huber [7], both of which suggest that the cross section decreases with decreasing energy. All three previous measurements relied on a hydrogen target created by dissociation of molecular hydrogen. This technique can lead to normalization problems (e.g., Ref. [4]). Furthermore, it is not known if the previous measurements had a significant fraction of metastables in their ion beams.

As can be seen in Fig. 2, the present MCLZ calculations for both the H and D targets show good agreement with our new measurements throughout the energy range considered. In particular, for collision energies less than 3 eV/u, the agreement is excellent. The lower energy measurements, taken with the D target, follow the energy dependence of the D target MCLZ calculations and are consistent with the significant isotope effect predicted for energies less than 5 eV/u. At higher energies, the MCLZ calculations reproduce the local peak at ∼50 eV/u, but the theoretical cross sections are ∼30% smaller. This discrepancy is likely related to the neglect of rotational coupling and short-range non-local interactions in the Landau-Zener approximation, both of which become important at the higher energies.

Due to the good agreement between the merged-beam total cross section measurements and the MCLZ calculations, it is reasonable and useful to report the calculated state-selective cross sections for the dominant channels. Fig. 3 displays the MCLZ H target state-selective cross sections for capture to the $2s^22p^23s$, $2s^22p^23p$, and $2s^22p^23d$ configurations of Ne^{3+}. Cross sections for capture to the $2s^22p^24s$, $2p^5\ ^2P^o$, and $2s2p^33s\ ^4S^o$ configurations are smaller than 5×10^{-17} cm^2 for the energy range considered and therefore not shown. Capture to the 3d dominates for collision energies less than 1000 eV/u with the 3p only becoming significant for higher energies. The five largest LS terms from the 3d configuration are also shown in Fig. 3. For collision energies greater than 2 eV/u, the dominant capture channel is the $2s^22p^2(^3P)3d\ ^4P$ which peaks near 50 eV/u, accounting for the local maximum in the experimental cross section. For lower energies, the 3d configuration cross section rises due to the increase in the $2s^22p^2(^1D)3d\ ^2F$, ^2D, and ^2P capture channels. This sharp increase in the cross section is attributed to trajectory effects due to the ion-induced dipole attraction between the reactants. The isotope effect is manifested in the MCLZ calculation purely in the 3d configuration. The ratio between the cross sections with H and D is predicted to be a factor of 1.3 at 0.01 eV/u.

V. CONCLUSIONS

Using a merged-beams technique, the absolute total electron capture cross section has been measured for Ne^{4+} on H(D) for the collision energy range of 0.10-1006 eV/u. The cross section above 250 eV/u was measured with H and below 250 eV/u was measured with D. Multichannel Landau-Zener calculations are performed with both H and D from $0.01 - 1000$ eV/u and compare well to the measured total cross sections. Below ∼5 eV/u, the present total cross section calculations show a significant target isotope effect. Agreement between the merged-beam measurements and the MCLZ predicted cross sections gives some confidence in the MCLZ predicted state-selective cross sections. For capture to the Ne^{3+}(3d) manifold, the ratio between the cross sections with H and D is predicted by the MCLZ calculation to be a factor of 1.3 at 0.01 eV/u.

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TABLE I: Ion-atom merged-beams cross section data for $Ne^{4+}+H(D)\rightarrow Ne^{3+}+H^+(D^+)$ as a function of collision energy. Measurements below(above) 250 eV/u are performed with D(H). Also listed is the statistical uncertainty and total combined (statistical plus systematic) uncertainty estimated at the 90% confidence level. See text for details.

Collision Energy (eV/u)	Cross Section (10^{-16} cm^2)	Statistical Uncertainity (10^{-16} cm^2)	Total Uncertainty (10^{-16} cm^2)
0.10 ± 0.02	36.7	6.6	8.0
0.17 ± 0.04	23.6	4.7	5.5
0.31 ± 0.05	20.2	6.4	6.9
0.50 ± 0.04	21.7	2.3	3.5
0.77 ± 0.04	16.5	1.9	2.8
1.11	23.5	3.7	4.6
1.79	20.2	2.3	3.4
2.02	19.7	2.4	3.3
2.69	20.2	3.2	4.0
3.20	25.6	2.6	4.0
4.28	26.6	3.2	4.5
6.50	30.5	3.4	4.9
10.2	29.8	1.3	3.8
15.2	35.7	3.6	5.5
24.2	38.9	1.8	5.0
32.0	38.0	2.4	5.8
48.0	44.5	2.6	5.9
70.4	41.2	2.5	5.6
104	41.2	3.3	6.0
145	38.2	2.3	5.2
195	35.7	2.9	5.2
286	36.3	2.2	4.8
371	37.3	1.9	4.8

Table I (cont'd)

403	37.4	2.6	5.2
471	34.1	2.1	4.5
548	36.8	2.2	4.9
621	35.3	1.4	4.4
857	36.6	1.5	4.6
1006	38.0	2.5	5.2

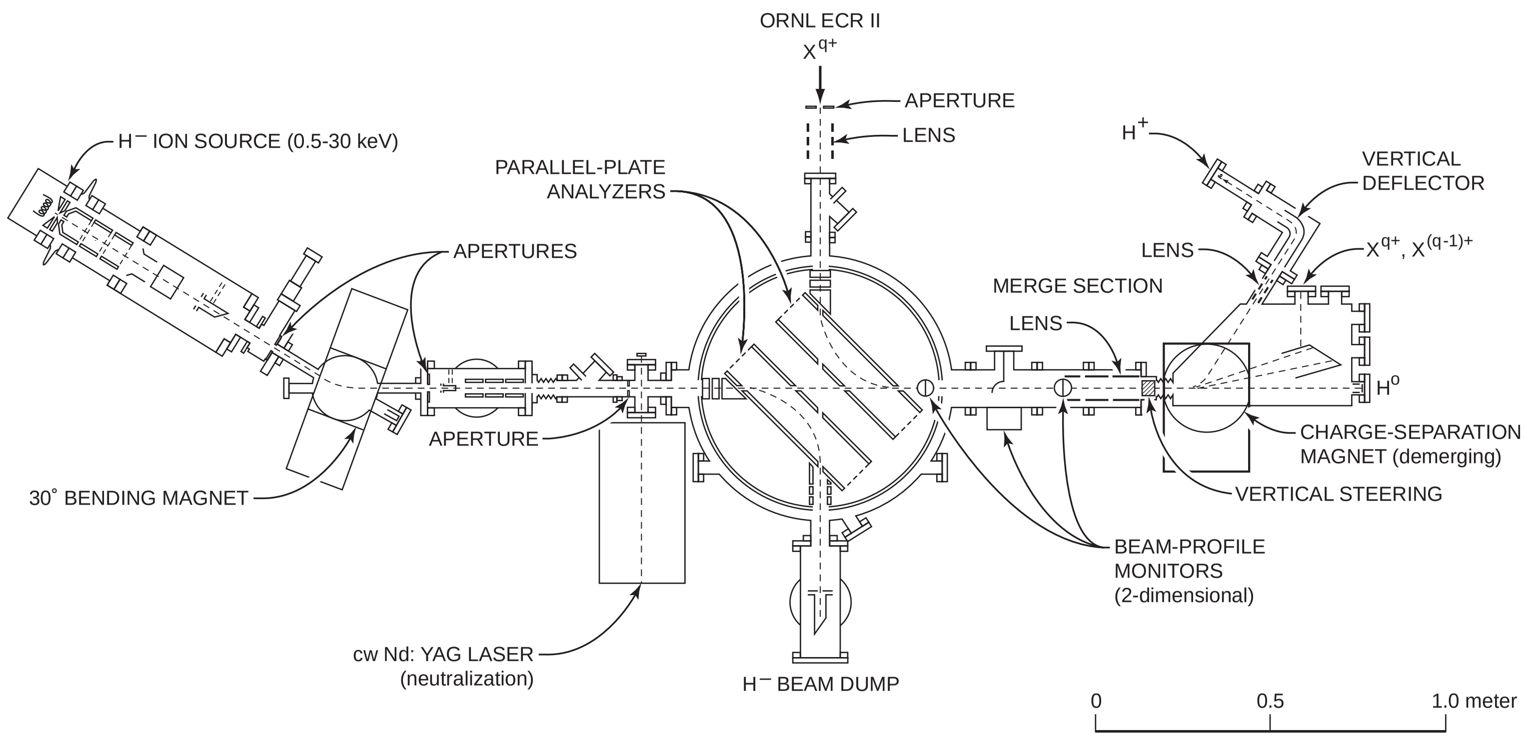

FIG. 1: Schematic of the ion-atom merged-beams apparatus

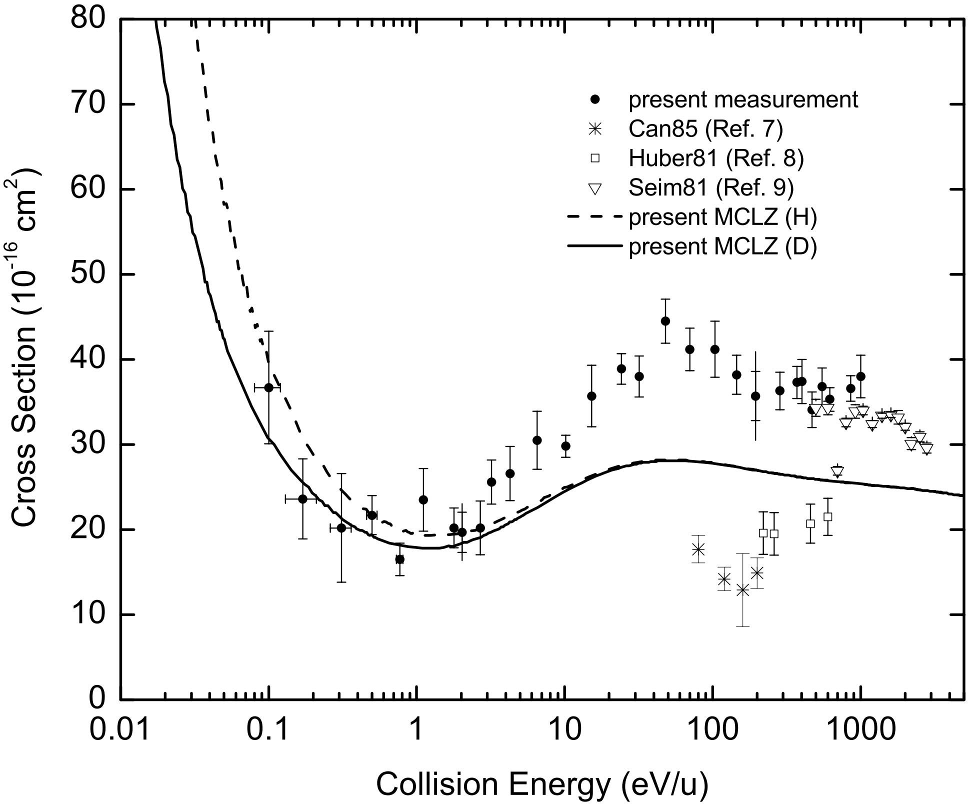

FIG. 2: Ion-atom merged-beam measurements and MCLZ calculations of the electron-capture cross section for Ne^{4+}+H(D) as a function of collision energy. Previous experiments are also shown. The statistical errors (estimated at a 90% confidence level) of the present measurements are shown. At energies 2.02 and 195 eV/u both the relative and total errors are shown at a 90% confidence level. For collision energies less than 1 eV/u the uncertainties in center-of-mass collision energy are also shown by horizontal lines. See text for details.

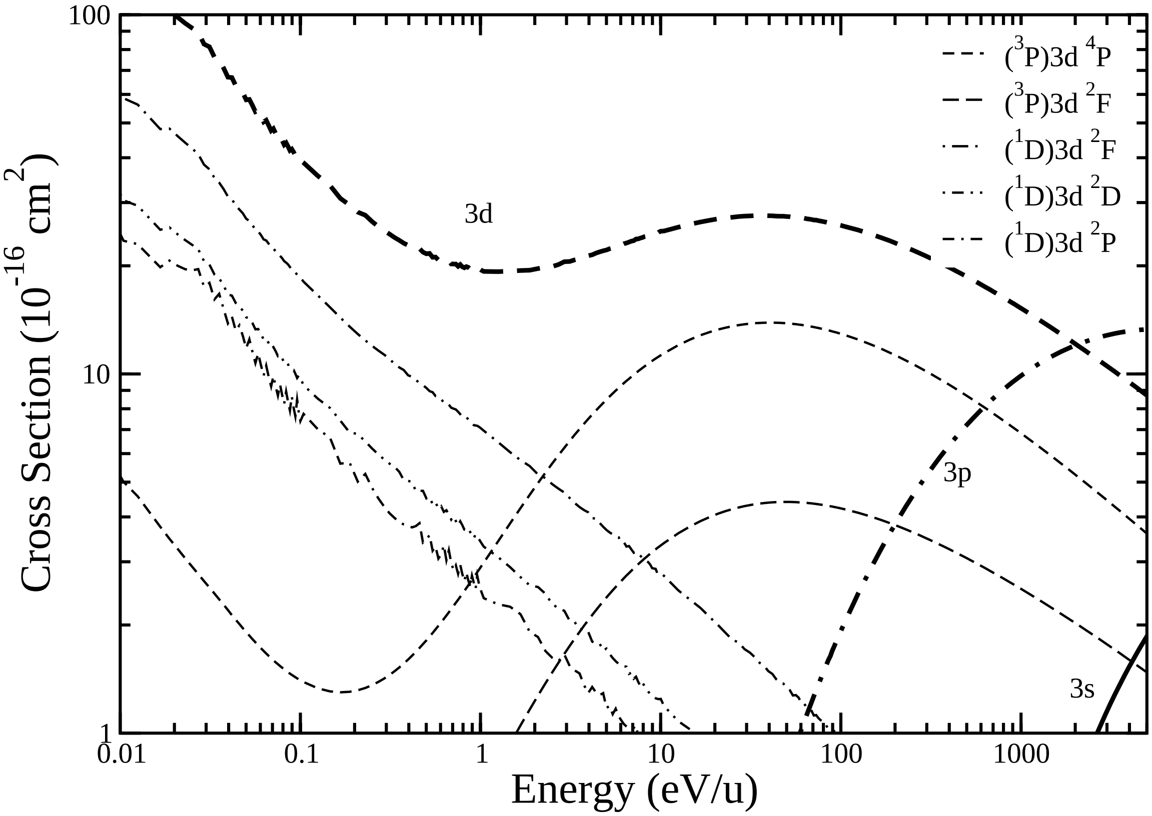

FIG. 3: State-selective MCLZ cross sections for Ne^{4+} + H → Ne^{3+} + H^{+}. Thick lines correspond to capture to the Ne^{3+} configurations $2s^22p^23s$, $2s^22p^23p$, and $2s^22p^23d$. Thin lines correspond to the indicated LS terms for the $2s^22p^23d$ configuration.