Electron capture by Ne²⁺ ions from atomic hydrogen

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Using a merged-beam technique, the absolute, total electron-capture cross section has been measured for collisions of Ne²⁺ ions with hydrogen (deuterium) atoms at collision energies between 139 and 1490 eV/u. These data are compared to three other published measurements, two of which differ from one another by a factor greater than two. Early quantal rate coefficient calculations for Ne²⁺ ions with hydrogen at eV/u energies predict a cross section many orders of magnitude below the previously measured cross section at 40 eV/u. A possible explanation is given for the discrepancy between theory and experiment.

I. INTRODUCTION

Electron capture from atomic hydrogen at low relative collision energies (meV/u–keV/u) is an important recombination mechanism in many plasmas, such as those found in fusion devices and in cosmic photoionized plasmas. Much theoretical and experimental work has been performed in order to provide the electron-capture (EC) cross-section data needed to model and interpret the spectra of these laboratory and astrophysical plasmas. However, as the need for more accurate data increases, previous measurements often show significant unresolved discrepancies. Fully quantum coupled-channel molecular-orbital calculations are considered the most accurate but are difficult to perform. Recent reviews of the relevant EC theory and experiment have been given by Stancil [1] and Havener [2], respectively. Throughout this paper, collision energies are quoted using the center-of-mass (c.m.) energy divided by the reduced mass of the colliding particles (i.e., eV/u).

In fusion energy research, EC cross sections at eV/u energies to hundreds of eV/u are needed for the accurate modeling and diagnostics of the scrape-off layer (i.e., edge) plasma. An issue of particular interest is the effect impurity ions have on fusion plasmas. For example, some of the effects of impurities have been studied by injecting Ne into DIII-D, the third generation tokamak developed by General Atomics in San Diego, CA. The subsequent radiation, as the highly charged Ne ions underwent EC with deuterium, reduced heat efflux while edge confinement degraded [3]. Accurately modeling the Ne charge balance and radiative cooling in these plasmas requires reliable EC cross-section values for a wide range of energies and Ne ionization stages.

In astrophysics, EC cross sections are important for calculating the ionization balance of planetary nebulae and H II regions. These calculations are used to interpret observations and to infer the chemical abundances of the observed

sources. EC rate coefficients for Ne ions in particular have been an issue of some concern since 1978 when Péquinot *et al.* [4] found that photoionization models failed to reproduce the observed spectrum of the planetary nebula NGC 7027. They suggested that inclusion of electron capture by Ne²⁺ with H would help to resolve the discrepancy. However, an early quantal coefficient calculation by Butler *et al.* [5,6] estimated that the EC cross section was far too small to resolve this discrepancy. The calculations were done in the plasma temperature range from 5000 K to 50 000 K and involved only transitions from the initial ground state $[\text{Ne}^{2+}(2s^2 2p^4 \, ^3P) + \text{H}]$ to the strongly exoergic final state $[\text{Ne}^+(2s^2 2p^5 \, ^2P) + \text{H}^+]$ considering only two relevant ²Π quasimolecular adiabatic states. These states, with a separated atom limit of 27.2 eV, are far apart at all internuclear distances. This results in a sharply decreasing cross section toward thermal energies. Further investigation by Forster *et al.* [7] considered initial metastable excited states of Ne²⁺ and slightly exoergic final states of Ne⁺, within the doublet manifold of adiabatic states, but still estimated that the EC rate coefficients are much too low to explain the values inferred from observations of planetary nebulae. Since that time, Ne physics has remained an issue of concern. For example, Pottasch and Beintema [8] have noted factor-of-3 discrepancies among the various Ne abundance determinations for the planetary nebula NGC 6302. Pottasch and Beintema suggested uncertainties in the underlying atomic data as one of the several possible causes for the discrepancies.

To address these issues, a series of measurements of the absolute EC cross sections of Ne^{q+} (q=2,3,4) with H(D) has been initiated. Results for Ne²⁺+H(D)→Ne⁺ +H⁺(D⁺) at collision energies 139–1490 eV/u are reported here. We are unaware of any calculation of the EC cross section for Ne²⁺+H at energies for which laboratory measurements have been carried out. Comparisons are made with the He²⁺+H system, which shows a similar energy dependence toward lower collision energies. We also discuss the possibility that transitions between quartet quasimolecular states of (NeH)²⁺, rather than between doublet states, might explain the measured data.

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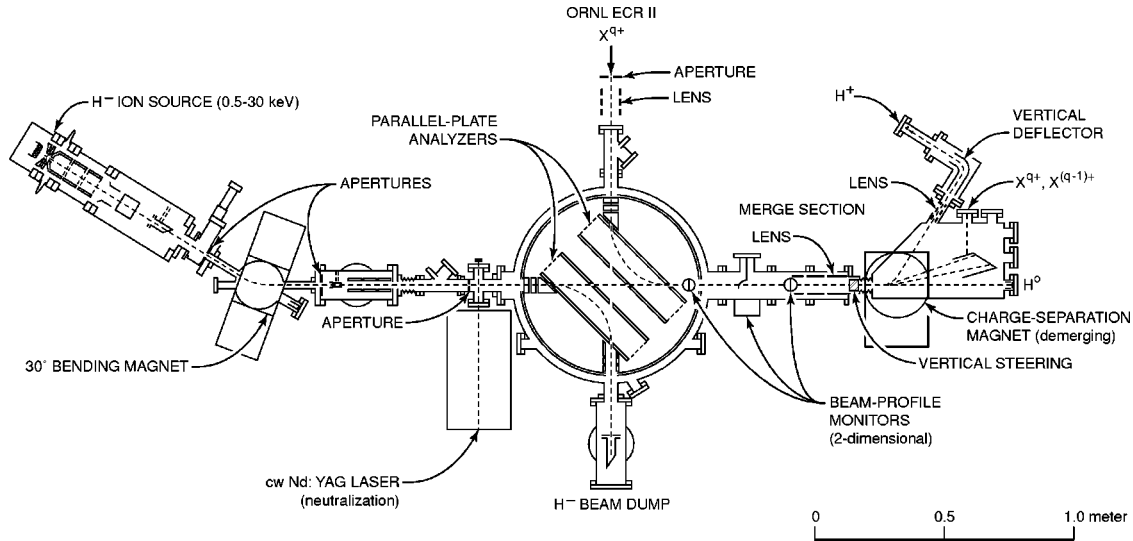[†]Electronic address: havenercc@ornl.gov

FIG. 1. Schematic of the ion-atom merged-beam apparatus.

The paper is organized as follows: In Sec. II the experimental setup used in measuring the EC cross section of Ne^{2+} ions on H(D) is described. The results are presented and discussed in Sec. III. Conclusions are given in Sec. IV.

II. EXPERIMENTAL APPROACH

The measurement of the EC cross section for the Ne^{2+} +H system was performed using the Oak Ridge National Laboratory (ORNL) ion-atom merged-beam apparatus, which has previously been described [9,10] in detail. The apparatus is depicted schematically in Fig. 1. In our setup, relatively fast (keV) beams are merged providing for a large dynamic range of collision energies in the c.m. [10], allowing access to collision energies from keV/u down to meV/u. In the present investigation, a Ne^{2+} beam with energies of 16–44 keV was merged with a faster D beam at energies of 6, 7, and 9 keV. This range of beam energies allowed for the total EC cross section to be measured in the energy range of 139–1490 eV/u. Lower c.m. collision energies can potentially be achieved by using either a slower D beam or a faster ion beam. However, operation of the D^- source at voltages lower than 6 kV or the ECR source at voltages higher than 22 kV led to poor quality or unstable beams.

The Ne^{2+} beam was produced by the ORNL CAPRICE ECR ion source [11] with an intensity of ≈ 4 μA, a diameter of 2–4 mm (full width at half maximum), and a (half-angle) divergence less than 0.25°. The cross section for capture onto an ion with an excited core could differ significantly from capture onto an ion in the ground state [12]. The possibility of metastables in the Ne^{2+} beam extracted from an ECR ion source was investigated by Bannister [13] through measurements of the electron-impact ionization cross section of the Ne^{2+} beam from the ORNL ECR source [14] using the electron crossed-beam apparatus [13]. No ionization signal was observed below the ground-state ionization threshold of 63.45 eV, indicating no detectable population of metastables in the Ne^{2+} beam.

A fast neutral ground-state D atom beam was obtained by

photodetachment of a D^- beam as it crossed the optical cavity of a 1.06-μm cw Nd:YAG (yttrium aluminum garnet) laser where kilowatts of continuous power circulate. The D^- beam was extracted from a duoplasmatron source. Collisional detachment of the D^- beam on background gas resulted in a small fraction (0.01%) of excited states in the D beam. The D beam obtained was nearly parallel (the divergence is less than 0.15°) with a beam diameter of 2 mm and intensities ranging from 10–20 nA. Deuterium was used instead of hydrogen to maximize the angular acceptance of the apparatus [9,15]. Isotope effects from the use of deuterium, due to differences in trajectories [16], are not expected at these energies. Such isotope effects are known to exist for ions of higher charge (e.g., Si^{4+}+H(D) [17]) at collision energies below 1.0 eV/u. Isotope effects above 100 eV/amu are only predicted for ions of charge greater than 10 [18].

As depicted in Fig. 1, the Ne^{2+} beam was electrostatically merged with the neutral D beam. Both beams interacted along a field-free region of 47 cm, after which the D^+ product ions were magnetically separated from the primary beams. The neutral beam was monitored by measuring secondary electron emission from a stainless steel plate and the intensity of the Ne^{2+} beam was measured using a biased Faraday cup. The product signal D^+ ions were detected by a channel electron multiplier. The signal rate (Hz) was extracted from background (kHz) by a two-beam modulation technique [9]. To correct the signal rate for the small fraction of excited D, the signal was measured with and without the laser on. The difference between the signals corresponded to the signal due to collisions with photodetached ground-state D atoms.

Absolute electron-capture cross sections were determined at each velocity from directly measurable parameters by the following formula:

$$\sigma = \frac{R \gamma q e^2 v_1 v_2}{I_1 I_2 v_r F}, \tag{1}$$

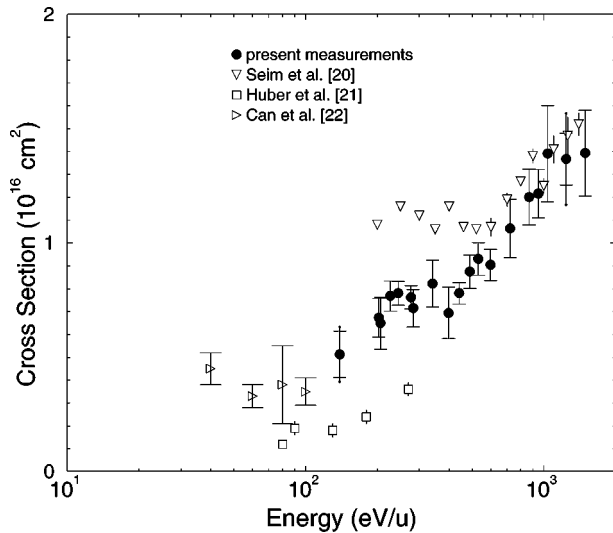where R is the signal count rate, q is the charge of ion, e is

FIG. 2. Present ion-atom merged-beam measurements of the electron-capture cross section for Ne^{2+}+H(D)→Ne$^+$+H$^+$(D$^+$) as a function of center-of-mass collision energy. A comparison is shown with other measurements. The statistical errors (estimated at a 90% confidence level) of the present measurements are shown, except at energies 139 eV/u and 1240 eV/u, where both the statistical and total errors are shown at a 90% confidence level. The reported statistical uncertainties of Seim *et al.* [20], Huber *et al.* [21], and Can *et al.* [22] are also shown. Not shown are their total experimental uncertainties.

the electronic charge, I_1 and I_2 are the intensities of the two beams, v_1 and v_2 are the velocities of the beams, v_r the relative velocity between beams, γ is the secondary electron emission of the neutral detector, and $\langle F \rangle$ is the average form factor which is a measure of the overlap of the beam. The form factor was estimated from two-dimensional measurements of the overlap at three different positions along the merge path. The secondary electron emission coefficient γ was measured *in situ* as described previously [9] and found to range from 1.32 ± 0.03 for 7-keV D$^-$ to 1.49 ± 0.03 for 9-keV D$^-$. For the 6-keV beam, γ was determined by linear extrapolation of γ for 7-keV and 9-keV beams [9]. The velocities were calculated from the energies of the beams, which included the estimated plasma potential shifts of the two sources (see, e.g., Ref. [17]). The relative merge angle between beams was negligible for these relative collision energies and assumed to be zero.

III. RESULTS AND DISCUSSION

Figure 2 shows the measured absolute EC cross section for the Ne^{2+}+D system as a function of c.m. collision energy. The error bars on the experimental data indicate the statistical error at a 90% confidence level. At energies of 139 eV/u and 1240 eV/u the total uncertainty is also shown. This total uncertainty is the quadrature sum of the statistical uncertainties and the 12% estimated systematic error [19]. Table I lists the data and includes both statistical and total errors.

TABLE I. Ion-atom merged-beam cross section data for Ne^{2+}+D→Ne$^+$+H$^+$(D$^+$) as a function of collision energy. Also listed is the statistical uncertainty and total combined (statistical plus systematic) uncertainty estimated at the 90% confidence level (C.L.).

Relative collision energy (eV/u)	Neutral beam energy (keV)	Cross section (10^{-16} cm^2)	Statistical uncertainty (90% C.L.) (10^{-16} cm^2)	Total absolute uncertainty (10^{-16} cm^2)
139	6.0	0.51	0.10	0.12
203	6.0	0.67	0.08	0.12
206	7.0	0.65	0.11	0.14
227	7.0	0.77	0.07	0.11
245	7.0	0.78	0.05	0.11
277	7.0	0.76	0.05	0.10
283	6.0	0.72	0.08	0.12
341	7.0	0.82	0.10	0.14
398	9.0	0.69	0.11	0.14
441	7.0	0.78	0.05	0.10
489	9.0	0.87	0.07	0.13
530	7.0	0.93	0.07	0.13
597	9.0	0.90	0.07	0.13
722	9.0	1.1	0.13	0.18
867	9.0	1.2	0.12	0.19
948	9.0	1.2	0.11	0.18
1040	9.0	1.4	0.22	0.27
1240	9.0	1.4	0.11	0.20
1490	9.0	1.4	0.19	0.25

As can be seen in Fig. 2, the present data agree with the results of Seim *et al.* [20] at energies above 700 eV/u, but are below their results at lower energies. Although the cause of this discrepancy is unclear, possibilities include uncertainties in the N^{q+}+H(q=2–5) EC cross section used to normalize their measurements and the incomplete dissociation of H$_2$ into atomic hydrogen inherent in their atomic source. Metastable contamination is not believed to be an issue. Seim *et al.* measured the metastable content of their Ne^{2+} beam and found it to be negligible. Furthermore, calculations at lower energies (eV/u) by Forster *et al.* [7] for EC with metastable Ne^{2+} indicate that the rate coefficient, while larger than the previous estimates for the ground state (1.0×10^{-20} cm^3 s^{-1}), is still negligible (6.0×10^{-14} cm^3 s^{-1}).

The Huber *et al.* [21] data are systematically lower than both our data and the data of Can *et al.* [22] (see Fig. 2). Together these comparisons suggest, as was first proposed by Can *et al.* [22], that a systematic error exists in the measurements of Huber *et al.* In their experimental procedures, both Huber *et al.* and Can *et al.* normalize their Ne^{2+}+H to their respective Ne^{2+}+H$_2$ results. Can *et al.* used their Ne^{2+}+H$_2$ results to renormalize the data of Huber et al., essentially multiplying these data by a factor of ≈ 2. This brought the data of Huber *et al.* into agreement with Can *et al.* This also brings the results of Huber *et al.* into agreement with our measurements. Although the energy range of our data
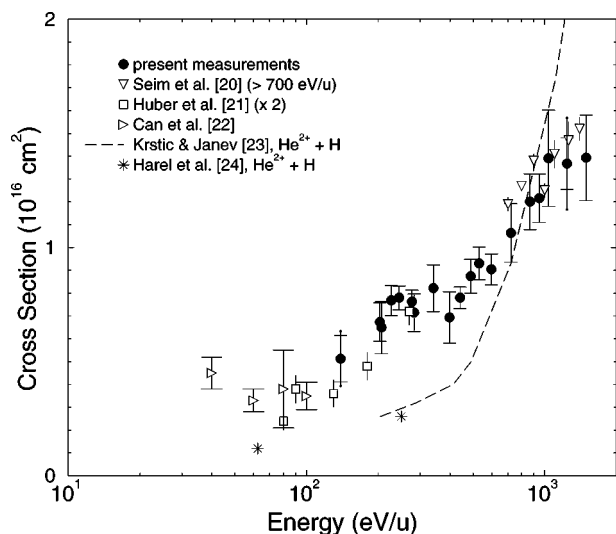
FIG. 3. Present ion-atom merged-beam measurements and corrected previous data of the electron-capture cross section for $Ne^{2+}+H(D)$ as a function of center-of-mass collision energy. Comparison is made with theoretical calculations for $He^{2+}+H$ by Krstic and Janev [23] and by Harel *et al.* [24]. See text for details.

does not overlap with that of Can *et al.* [22], our results do appear to map relatively smoothly into theirs.

Figure 3 shows a "recommended" experimental cross section for $Ne^{2+}+H$ which includes our present measurements and the above corrections to the previous data. The Huber *et al.* data has been multiplied by a factor of 2 and only the Seim *et al.* data above 700 eV/u are included in the figure. Figure 3 also shows a comparison with two theoretical calculations for $He^{2+}+H$ [23,24], a collision system with the same incident charge but without electrons on the ion core. Any observed differences (or similarities), then, are due to interactions with the multielectron, open-shell Ne^{2+} core. Such a comparison was previously done [25] for $Cl^{7+}+H$ measurements and $N^{7+}+H$ calculations from a few eV/u to several hundred eV/u. For this case the closed Ne-like shell of Cl^{7+} did not significantly influence the EC dynamics leading to practically identical cross sections for $N^{7+}+H$ and $Cl^{7+}+H$.

As shown in Fig. 3, both the $Ne^{2+}+H$ and the $He^{2+}+H$ collision systems show a sharply decreasing cross section from keV/u to ~400 eV/u. Below ~400 eV/u, the cross sections deviate from this decline, showing a change in slope. Motivated by this similarity in shape of the theoretical EC cross-section curves for $He^{2+}+H$ and the present measurement for $Ne^{2+}+H$ system, a qualitative analysis was performed to identify possible EC channels that are present in both systems. Figure 4 presents the relevant adiabatic quasimolecular electronic potential-energy curves for (a) $(NeH)^{2+}$ and (b) $(HeH)^{2+}$ as functions of internuclear distance. The curves for the $(NeH)^{2+}$ [Fig. 4(a)] were calculated using full configuration interaction in the space of 42 Gaussian basis functions (S,L,P), using the computational chemistry code GAMESS [26], while the $(HeH)^{2+}$ curves
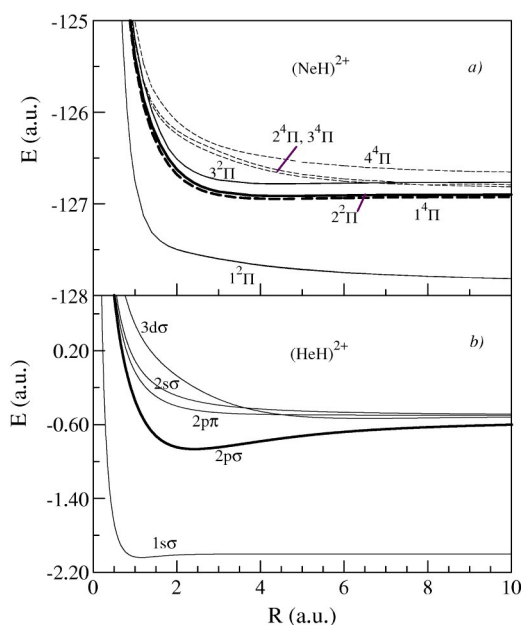

FIG. 4. Adiabatic potential-energy curves for (a) $(NeH)^{2+}$ and (b) $(HeH)^{2+}$ as functions of internuclear distance. In (a) the initial Ne^{2+} ^3P+H channel corresponds to the $2\,^2\Pi$ (thick solid line) and $1\,^4\Pi$ (thick dotted line) quasimolecular states. In (b) the initial $He^{2+}+H(1s)$ channel corresponds to the $2p\sigma$ quasimolecular state (heavy solid lines). (See text for details.)

[Fig. 4(b)] were generated by the standard closed-form two-center, one-electron algorithm [23].

Considering first the $Ne^{2+}+H$ collision system, as the reactants approach each other, the ground ^3P state of Ne^{2+} combines with the electron spin of the hydrogen forming either spin doublet or quartet quasimolecular states. These states are degenerate at large internuclear distances. Discarding the weak spin-orbit interaction, the collision system evolves independently along these two noninteracting spin configurations, i.e., the $2\,^2\Pi$ and $1\,^4\Pi$ adiabatic states of $Ne^{2+}+H$ [thick solid and thick dashed lines in Fig. 4(a)]. These states are initially populated according to doublet (2) and quartet (4) spin statistics (neither projectile nor target are spin polarized). The $2\,^2\Pi$ and $1\,^4\Pi$ couple to other doublet and quartet adiabatic states, respectively, and the EC cross section is made up of separate contributions of the doublet and quartet manifolds of states of $(NeH)^{2+}$. A set of the lowest adiabatic doublet (thin solid lines) $^2\Pi$ and quartet (thin dashed lines) $^4\Pi$ states of Ne^++H^+ is shown in Fig. 4(a) as functions of internuclear distance R. The only electron-capture channel that is strongly exoergic and thus possible at sub-eV collision energies is from the initial doublet $2\,^2\Pi$ to the $1\,^2\Pi$ state. The $1\,^2\Pi$ state corresponds to the $Ne^+(2s^22p^5\,^2P)+H^+$ capture channel. Due to the large energy splitting, this channel is weak, resulting in a very small cross section [5]. Transitions to other doublet states as well as from $1\,^4\Pi$ to the quartet states shown in Fig. 4(a) are slightly endoergic.

Considering now the $He^{2+}+H$ collision system, the initial channel evolves along the $2p\sigma$ quasimolecular state [thick solid line in Fig. 4(b)]. A strongly exoergic EC product
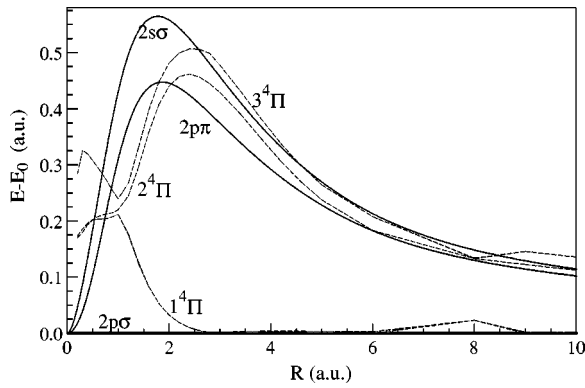
FIG. 5. Adiabatic electronic energy terms for $^4\Pi$ manifold of states of (NeH)$^{2+}$ close to the initial 1 $^4\Pi$ (thin dashed line), and (HeH)$^{2+}$ terms, relative to the $2p\sigma$ electronic term of (HeH)$^{2+}$ system. The $2p\sigma$ term coincides with the horizontal coordinate axis.

channel $1s\sigma$ is also present here, although the transitions to $2p\pi$, $2s\sigma$, and $3d\sigma$ states, which are at large distances exactly degenerate with the initial $2p\sigma$ state, dominate the cross section even at low energies. The change of slope in the EC cross section for He^{2+}+H system below keV/u energies in Fig. 3 is due to the strong rotational (Coriolis) coupling between $2p\sigma$ and $2p\pi$ states [23]. Unlike the dominant, tunneling mechanism for radial transitions, which results in exponential decrease, $\exp(-C/v)$, of the cross section toward lower energies [23] (v is the internuclear velocity and C is a constant relating to the Massey parameters), the decrease of the Coriolis cross section is slower, following the power law ($v^{2/3}$ for a straight line trajectory [27]). Similar mechanisms for EC capture might be present for the Ne^{2+}+H system, between quartet states that were not analyzed before and which might cause the apparent change of the slope in the EC cross section in the hundreds of eV/u range (as observed in the measurements, see Fig. 3). It is important to note that the lowest-energy calculation of Harel *et al.* [24] may exaggerate the contribution due to rotational coupling due to the use of straight-line trajectories, as indicated by recent fully quantal calculations of Liu *et al.* [28]. These calculations show that the cross section decreases again at lower energies due to the effects of nuclear repulsion on rotational transitions.

To investigate further, in Fig. 5 the same electronic adiabatic terms for (HeH)$^{2+}$ and (NeH)$^{2+}$ systems are shown. However, they are now minus the nuclear-repulsive potentials Z/R [$Z=2$ for (HeH)$^{2+}$ and $Z=10$ for (NeH)$^{2+}$] and choosing the ground, $2p\sigma$ curve of (HeH)$^{2+}$ for the zero energy (at each R). The degeneration of the 1 $^4\Pi$, 2 $^4\Pi$, and 3 $^4\Pi$ terms of (NeH)$^{2+}$, necessary for the strong rotational coupling (between substates of different magnetic number), is apparent in the united atom limit (~ 1 a.u.). Similarly, there is a degeneration of the $2p\sigma$, $2p\pi$, and $2s\sigma$ curves of the (HeH)$^{2+}$, indicating contribution to EC from the Coriolis $2p\sigma$-$2p\pi$ transition cross section. Since the main contribution to the Coriolis cross section in (HeH)$^{2+}$ comes from an extended range of internuclear distances of up to 2 a.u., the deformation of the terms inside the Ne$^+$ core (1 a.u.) is

not expected to significantly change the nature of the rotational coupling. The lower final quartet states of Ne$^+$+H$^+$ are only slightly endoergic with the initial channel (of the order of a fraction of an eV). Therefore, both the Coriolis and radial transitions from the initial 1$^4\Pi$ to the manifold of quartet states are energetically allowed at the energy range considered here and qualitatively explain the similarity between the EC cross sections for He^{2+}+H and Ne^{2+}+H systems. While the measured low-energy EC cross sections seem to have the energy dependence suggestive of the discussed coupling between quartets, it is not known how much rotational coupling will contribute at lower (above threshold) energies, where it is reduced because of the nuclear repulsion. The low-energy behavior of the He^{2+}+H cross section is currently being both theoretically and experimentally investigated at ORNL.

IV. CONCLUSIONS

Using a merged-beam setup, we have measured independent, absolute, total electron-capture cross sections for Ne^{2+} on H(D) for the collision energy range of 139–1490 eV/u. Good agreement with the cross sections measured by Seim *et al.* [20] is observed at energies above 700 eV/u. Our results drop with decreasing energy and are consistent with the low-energy measurements of Can *et al.* [22]. The present results are larger than the measurements of Huber *et al.* [21] by a factor of ≈ 2. A consistent set of experimental data is presented for energies between 40 eV/u and 2000 eV/u. We are unaware of any theoretical calculations of the EC cross section that overlap with the energy range for which measurements exist. Comparison is made with the He^{2+}+H collision system which shows a similar energy dependence toward lower energies. By comparing the electronic potential-energy curves for the two systems the flattening out of the cross section for Ne^{2+} is found probably due to rotational coupling to slightly endoergic quartet states. These states were not considered in previous estimates and may lead to a slight increase in the EC cross section at above threshold energies. An upgrade of the ORNL ECR facility is in progress which will provide the higher-energy ion beams needed to extend the ion-atom merged-beam measurements to energies below 100 eV/u where the cross section is expected to again decrease.

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