Excitation of atomic hydrogen by fully stripped ions

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Excitation of atomic hydrogen in initial quantum levels $n_i = 1,2,3$ colliding with multiply charged ions with charge states q from 1 to 26 is investigated in the impact energy range 10 (keV/u)/n_i^2–10 MeV/u, by means of the classical trajectory Monte Carlo, many-state atomic orbital close-coupling, and symmetric eikonal formalisms. This extensive compilation of theoretical calculations confirms the feasibility of an empirical scaling relation (σ/q vs E/q) proposed in previous works to reduce the excitation cross sections induced by multiply charged ions to a single universal curve. This scaling, together with the semiempirical formula derived by Lodge and co-workers [J. Phys. B **9**, 239 (1976)] for proton projectiles, is found to provide reliable excitation cross sections for the one-electron collision system. Good agreement is obtained between theory and experiment for proton impact on H(1s) at impact energies above 10 keV.

I. INTRODUCTION

The theoretical study of the collisional excitation of atomic hydrogen by fast protons has been the object of numerous works in the last decades. For proton impact, these works have shown that perturbative approximations provide reliable results for impact velocities (v) greater than the mean velocity of the electron in its initial state (v_i). However, as v approaches v_i, more rigorous theoretical models are required in order to account for the strong coupling between the excitation, ionization, and electron-capture reaction channels. This region of impact velocities where all inelastic channels become strongly coupled is usually known as the intermediate impact energy regime. For singly charged projectiles, the intermediate velocity region is approximately within the interval $0.5 \lesssim v/v_i \lesssim 2$, where for a hydrogen target $v_i = 1/n_i$. For impact velocities below this range the ionization channel becomes negligible. On the other hand, for impact velocities above this range, the electron-capture channel becomes unimportant.

For multiply-charged-ion impact, the boundaries of the intermediate velocity region depend upon the projectile charge (q) and, in general, this region becomes wider for increasing projectile charges. Thus, extreme care must be taken in applying theoretical methods at impact energies that are usually considered to be above the intermediate energy region for single charged projectiles. This is probably the reason why very few attempts have been made to predict the excitation cross sections of hydrogen induced by multiply charged heavy ions. In contrast, there is a considerable need for this kind of information in order to be able to assess the parameters and the feasibility of neutral beam heating and current drive of the next generation of tokamak fusion reactors.[1]

Theoretical work on the excitation of hydrogen by multiply charged ions has been recently conducted by Olson[2] and Reinhold, Falcon, and Miraglia[3] using the classical trajectory Monte Carlo method (CTMC), Janev and Presnyakov[4] with a simplified three-state model, Fritsch and Schartner[5] with a many-state atomic orbital (AO) close-coupling formalism, Ryufuku[6] by means of a unitarized distorted wave approach, and Reinhold and coworkers[3,7] and Rodriguez and Miraglia[8] using a symmetric eikonal (SE) approximation.

On the other hand, experimental studies on excitation of hydrogen have been performed only for the case of proton projectiles. These are the energy loss measurements of Park *et al.*[9] for excitation of H(1s) to the levels $n = 2,3,4$ in the impact energy range 15–200 keV, the spectroscopic measurements of Morgan, Geddes, and Gilbody[10] for excitation of H(1s) into the 2s and 2p states in the impact energy range 2.3–26 keV, and the recent spectroscopic measurements of Schartner, Detleffson, and Sommer[11] for excitation of H(1s) into np states at impact energies from 80 to 700 keV.

In this work we extend previous studies of the excitation of hydrogen by multiply charged ions to consider projectile charge states from $q = 1$ to 26, and hydrogen targets in either the ground state or excited levels. That is, we shall be concerned with the reactions

$$A^{q+} + \mathrm{H}(n_i) \rightarrow A^{q+} + \mathrm{H}(n_f) \tag{1}$$

where n_i and n_f denote the initial and final quantum levels, respectively.

Our theoretical work consists of the application of the CTMC, AO, and SE formalisms to the study of reactions (1) for excitation from initial quantum levels $n_i = 1,2,3$ to final levels $n_f = (n_i+1),(n_i+2),(n_i+3)$. The impact energy range investigated is 10 (keV/u)/n_i^2 to 10 MeV/u,

which includes the difficult intermediate impact energy range. As with any theoretical approach, our models have inherent limitations. Thus, we have devoted Sec. II of this work to discuss these limitations in order to estimate the validity range of each of our formalisms. In Sec. III we present and discuss the result of our extensive theoretical calculations. Whenever possible, these calculations are compared with the available experimental data.

We analyze the possibility of reducing the excitation cross sections for different charge states of the projectile to a common plot. This is done by displaying the cross section divided by the projectile charge as a function of the impact energy in keV/u divided the projectile charge [i.e., a σ/q versus E (keV/u)/q plot]. This convenient plot was first introduced by Olson *et al.*[12] who demonstrated that a large compilation of experimental and theoretical multiply-charged-ion electron loss cross sections can be approximately reduced to a single curve. The feasibility of this kind of plot for excitation cross sections was first analyzed by Janev and Presnyakov[4] with an approximate three-state model and confirmed later by Fritsch and Schartner[5] using more realistic many-state calculations. Recent measurements by Reymann *et al.*[13] have confirmed the utility of such a plot for excitation of helium by multiply charged ions. Atomic units will be used throughout except where otherwise stated.

II. THEORETICAL METHODS

We shall be concerned with the scattering of structureless projectiles with impact velocity v (impact energy E) and charge states q by a hydrogen atom in a quantum state consisting of an equally populated combination of bound states with the same principal quantum number n_i. We will be interested in studying the excitation cross section from this initial quantum level to a final level determined by the principal quantum number $n_f > n_i$, i.e., reaction (1). Formally, the statistically averaged cross section for this process is given by

$$\sigma(n_i \rightarrow n_f) = n_i^{-2} \sum_{\substack{l_f, m_f \\ l_i, m_i}} \sigma(n_i, l_i, m_i \rightarrow n_f, l_f, m_f) \qquad (2)$$

where $\sigma(n_i, l_i, m_i \rightarrow n_f, l_f m_f)$ is the excitation cross section from the hydrogenic state determined by the quantum numbers (n_i, l_i, m_i) to the state determined by (n_f, l_f, m_f).

Deexcitation cross sections can be easily obtained from the detailed balance relation

$$\sigma(n_f \rightarrow n_i) = \frac{E_i}{E_f} \frac{n_i^2}{n_f^2} \sigma(n_i \rightarrow n_f) \qquad (3)$$

where E_i and E_f are the initial and final kinetic energy of the projectile, respectively.

A. The classical trajectory Monte Carlo method

The CTMC method has been successfully and extensively used for more than two decades in ion-atom collisions.[14,15] This method is well known for providing reliable total capture and ionization cross sections in the intermediate impact energy region in collisions involving multiply charged ions. However, very few attempts have been made to study target excitation processes.[2,3,16]

So far, several CTMC models for the hydrogen target have been proposed which differ in the choice of the initial phase-space electronic state. In this work we have adopted the microcanonical probability density proposed by Abrines and Percival.[14] A very important property of this kind of distribution is that it reproduces exactly the momentum distribution of an equally populated combination of all the substates of a given initial quantum level.[17,18] Therefore, the averaging over the initial states in Eq. (2) is implicitly included in the calculation and there is no need for independent runs to compute the state-to-state cross sections $\sigma(n_i, l_i, m_i \rightarrow n_f, l_f, m_f)$.

Because the final binding energies obtained with the CTMC method are continuous, there are problems in the definition of the transition between quantum levels.[19] These problems lie in the assignment of a definite band of binding energies to a given quantum level. The solution to this is not unique and induces problems with detailed balance relations. In this work we have adopted the solution proposed by Becker and MacKellar[20] which satisfies a principle of proportionality of classical and quantal weights. Following these authors, a range of final binding energies E_f of the electron is associated with a quantum n_f level according to the relation

$$[(n_f - 1)(n_f - 0.5)n_f] < (2E_f)^{-3/2} < [n_f(n_f + 0.5)(n_f + 1)] . \qquad (4)$$

Thus, even though classical mechanics is time reversible, it is not expected that the present model will satisfy exactly the detailed balance relation (3) due to the differences in the description of the initial and final states. As has been shown by Percival and Richards,[19] this problem disappears only for large values of n_i and n_f. In principle, departures from detailed balance for small values of n_i and n_f could be minimized if a band of binding energies is also associated with the initial phase-space state. However, this kind of extension of the present model goes beyond the scope of this article.

In spite of the problems in the definition of the initial and final states, the CTMC method has two very important properties which have been responsible for its success at intermediate energies: (i) all the interactions are exactly taken into account (i.e. none is considered as a perturbation) and (ii) all the possible reaction channels are considered simultaneously along with the coupling between them. These two properties, which become very important for increasing projectile charges, along with its computational feasibility, makes the CTMC method a very good candidate to study the behavior of the excitation cross sections produced by multiply charged ions at intermediate impact energies.

Semiempirically, the CTMC method is expected to provide reliable cross sections for proton projectiles in the approximate impact velocity range $1 < v/v_i < 4$.

Above this impact velocity range, the CTMC fails in predicting properly some optically allowed transitions.[19] As is well known, while the high-energy behavior of the CTMC dipole allowed excitation and ionization cross sections is proportional to $1/E$, the correct quantum behavior is proportional to $[\text{const}+\ln(E)]/E$.

For increasing projectile charges, the CTMC method is known to hold for higher impact velocities than for proton projectiles. In this work we will present CTMC excitation cross sections for H^+, He^{2+}, C^{6+}, O^{8+}, and Fe^{26+} projectiles in the impact velocity range $2<v/v_i<8.9$ (i.e., impact energies 100 keV/u $<En_i^2<2000$ keV/u). Therefore, we expect the CTMC cross sections to be valid in this range, except for the case of H^+ and He^{2+} projectiles at high impact energies, where the CTMC method should underestimate the true cross sections.

B. The symmetric eikonal approximation

The SE approximation is a distorted wave method that considers initial and final wave functions which are distorted by Coulombic eikonal phases. As has been shown in recent works,[7,21,22] this model provides reliable cross sections in the intermediate impact energy range. On the other hand, SE excitation cross sections tend to first Born cross sections at high impact energies, which is well known to be the correct high-energy limit for direct processes. Furthermore, previous calculations[3,7,23] have shown that the SE approximation predicts a dependence of the excitation cross section on the projectile charge which is in very good agreement with different experimental measurements for Fe^{24+}, F^{8+}, and Ca^{19+} ionic targets.

Because of the perturbative nature of the SE approximation, its validity for a given impact energy is determined by the magnitude of the projectile charge. Comparisons with experiments for the ionic targets mentioned above have indicated that the higher the impact energy, the larger the values of q for which the method holds. In this work we will present SE excitation cross sections for proton and completely stripped iron projectiles. For proton impact of hydrogen ($q=1$), we expect the applicability range of SE to be approximately $E>(15\text{ keV}/n_i^2)$. On the other hand, we estimate that, for iron projectiles ($q=26$), this model should provide reliable cross sections for impact energies $E>(400\text{ keV/u})/n_i^2$.

C. The atomic orbital close-coupling formalism

The semiclassical AO close-coupling formalism is one of the most rigorous theoretical approaches to deal with intermediate energy collisions. It considers all couplings among the quantum mechanical electron states that are included in a given basis set, including pseudostates which represent ionization channels and tighter binding in close collisions.[24,25] In principle, the convergence of calculated cross sections can be studied systematically as a function of the basis size if the basis is extended appropriately, e.g., as suggested by the Sturmian series. In the study of electronic excitation, a very large number of states would be needed to arrive at converged state-to-state cross sections due to the considerable mixing within the space of excitation and ionization states. In the present case, another adverse aspect of the numerical calculations consists of the need to consider very large impact parameters and collision times (proportional to q and n_i^2). Fortunately, the number of independent runs can be reduced (for $n_i>1$) by computing all the state-to-state cross sections from states within the same initial level n_i simultaneously.

In this work, we have chosen a one-center AO basis set of 74 states consisting of all the bound states of the levels $n=1$ to 5 of hydrogen (35 states of positive reflection symmetry) plus 39 pseudostates to represent the continuum of the target. For excitation from excited levels, the analogous states of negative reflection symmetry have also been included in separate calculations. This AO model should provide reliable cross sections only for those cases in which the most important reaction channels consist of direct processes, i.e., excitation with some coupling to the continuum of the target. Based on experiments and available two-center calculations for $H^+ + H(1s)$ collisions, we estimate the present calculations to be appropriate at energies above some 60 $[(\text{keV/u})/q]/n_i^2$. At lower energies, the lack of states centered in the projectile limits the validity of the present calculations.

III. RESULTS AND DISCUSSION

In Fig. 1 we compare the result of our calculations for the excitation cross sections from $H(1s)$ with all the available experimental data for proton projectiles. Very good agreement is observed between theory and experiment for excitation into $n_f=2$ at impact energies greater than 10 keV. Below this energy range, this figure clearly illustrates that molecular effects directly influence the cross sections.

For higher levels of excitation, the SE and AO calculations agree with each other whereas the CTMC cross sections become somewhat smaller. On the other hand, even though the uncertainties of the experimental measurements of Park *et al.*[9] are rather large, these data are still above the calculated cross sections.

Due to the lack of experimental data at impact energies above 200 keV, the calculations are compared with the recent measurements of the excitation cross sections into n_fp states of Schartner, Detleffson, and Sommer,[11] which should provide the major contribution to the total cross section at high energies. As expected, these experimental data are slightly smaller than the SE total excitation cross sections. In fact, we have verified that an excellent agreement is obtained between the SE cross sections for excitation into the n_fp subshell and the data of Schartner, Detleffson, and Sommer.[11]

In order to illustrate the complexity of the study of the excitation cross sections for multiply charged projectiles, we analyze in Fig. 2 the dependence of the calculated excitation cross section from $n_i=1$ to $n_f=2$ on the projectile charge at a fixed impact energy of 500 keV/u (i.e., at a fixed impact velocity of 4.47 a.u.). This impact energy is considered to be a high impact energy for proton projectiles and, therefore, the first Born approximation pro-

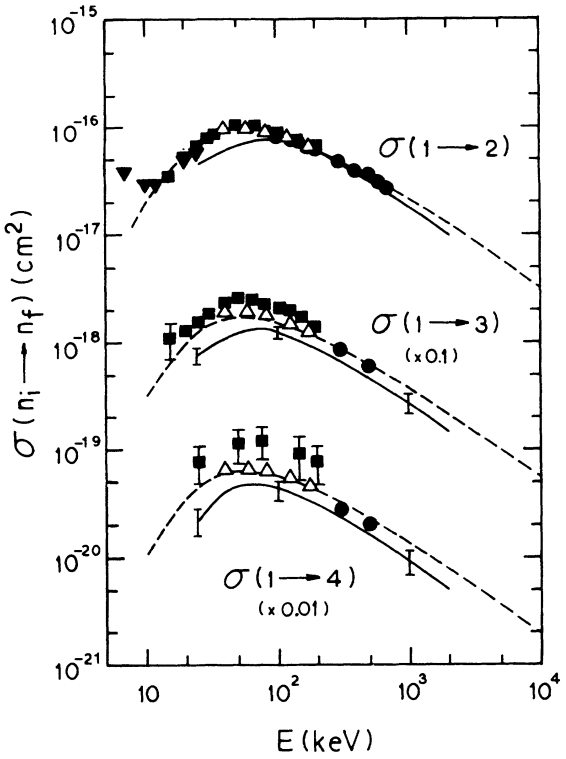

FIG. 1. Total excitation cross section in $H^+ + H(n_i=1) \rightarrow H^+ + H(n_f=2,3,4)$ collisions as a function of the impact energy. Experiments: ■, Park *et al.* (Ref. 9); ▼, Morgan, Geddes, and Gilbody (Ref. 10); ●, Schartner, Detleffsen, and Sommer (Ref. 11) for excitation to $n_f p$ levels. Theory: ——, present CTMC (error bars indicate statistical uncertainties); △, present AO; — — —, present SE.

vides reliable cross sections. However, departures from the q^2 dependence predicted by this simple first order theory are observed in the results of the other theories for increasing projectile charges. That is, this impact energy cannot be considered to be a high impact energy for large projectile charges. On the other hand, CTMC, AO, and SE, which go beyond first-order theories, essentially agree among themselves.

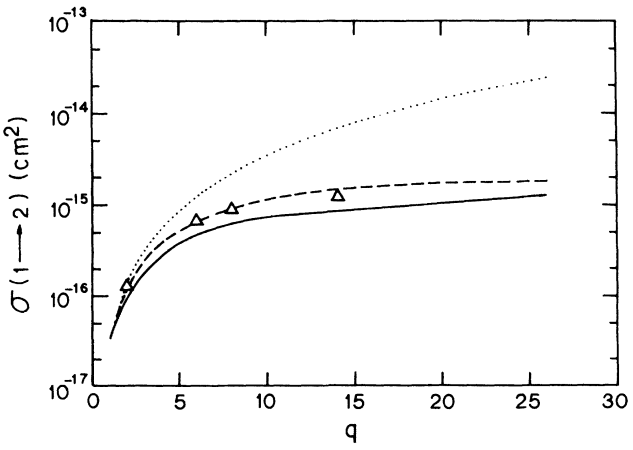

FIG. 2. Total cross section for excitation of hydrogen from $n_i=1$ to $n_f=2$ at an impact energy of 500 keV/u and as a function of the projectile charge. ——, CTMC; △, AO; — — —, SE; · · · ·, first Born approximation.

For increasing values of the projectile charge, our calculations present a plateau where the excitation cross sections become nearly independent of the projectile charge. This kind of behavior was termed "saturation of the cross sections" by Brendle *et al.*[26] who state that the excitation cross sections should tend to a constant value as the projectile charge tends to infinity. Similar behaviors were reported later by Wohrer *et al.*,[27] Reymann *et al.*,[13] Xu *et al.*,[28] Reinhold and co-workers,[3,7] and Rodriguez and Miraglia.[8,23]

Even though the study of the transitions produced by multiply charged ions at intermediate impact energies is rather complicated, it has been proposed by Janev and Presnyakov[4] and verified by Fritsch and Schartner[5] and Rodriguez and Miraglia[8] that the excitation cross sections from H(1*s*) to *p* states can be approximately reduced to a single curve. In this work, the corresponding curve would be

$$\sigma(E,q,n_i \rightarrow n_f) = qf(E/q,n_i,n_f) \tag{5}$$

where E is the impact energy in keV/u, q is the projectile charge, and f is a function that depends only on E/q.

As was found by Janev and Presnyakov,[4] the scaling relation (5) is satisfied exactly in their simplified three-state model. However, this model may be questioned[5] on the grounds that it neglects some important intercouplings. In fact, while the scaling relation was found empirically to be valid (except for $q=1$) in a many-state formalism, it was demonstrated to be severely violated in a consistent three-state formalism.[5]

In general, there is no obvious explanation of the scaling relation (5) since this scaling is not satisfied exactly by either the quantum mechanical or the classical equations that determine the evolution of the electronic wave function (i.e., either the Schroedinger equation or the classical Liouville equation). Therefore, the validity of this scaling has to be confirmed empirically, as was done previously for electron loss reactions.[12]

Nevertheless, simple classical[5,12] and quantum-mechanical[8] explanations of the scaling relation (5) were given in previous works for large projectile charges, where the major contribution to the excitation cross sections arises from large impact parameters. As these explanations become less valid for small projectile charges, departures from this scaling should be expected when the projectile charge changes from $q=1$ to 26, and indeed this has clearly been observed previously.[5,8]

In Figs. 3, 4, and 5 we analyze the validity of the scaling relation (5) for the excitation cross sections from the initial levels $n_i=1$, 2, and 3, respectively. Unfortunately, there exist no experimental data for projectile charges greater than 1. Therefore, our conclusions will be based on the results obtained with our three independent theoretical approaches.

In general, a good agreement is observed among the different theories. However, some discrepancies are observed as to the magnitude of the cross sections at the low and high scaled energy regions presented in the

figures. As explained in Sec. II A, the reason for the discrepancies between the CTMC and SE methods at high energies is the inability of CTMC to describe properly some of the dipole transitions.

On the other hand, at low scaled energies the capture channels and/or the production of fast free electrons becomes increasingly important and the single-center AO formalism may be considered less valid. Also, the excitation cross sections obtained with our SE method at scaled energies $E/q < 15$ keV/u and $n_i = 2,3$ are much smaller than the cross sections obtained with CTMC, particularly for $n_i = 3$ (Fig. 5) where SE and CTMC predict even different positions for the cross-section maxima. Here, since the CTMC method contains a more complete description of the couplings between all the reaction channels than SE, our results may indicate that this low-energy range could be beyond the validity of SE.

Concerning the scaling relation (5), all theoretical approaches confirm its approximate validity. However, some discrepancies are observed as to the magnitude of departures from this scaling. While the CTMC method predicts noticeable departures only for excitation from the ground state and at low scaled energies, appreciable departures are predicted by the AO and SE calculations

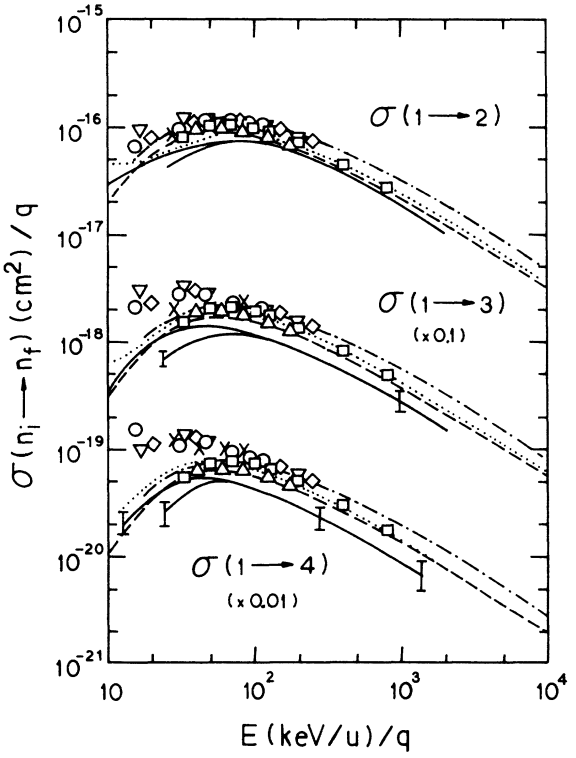

FIG. 3. Scaled total excitation cross section in $A^{q+}+\mathrm{H}(n_i=1) \rightarrow A^{q+}+\mathrm{H}(n_f=2,3,4)$ collisions as a function of the scaled impact energy. ——, present CTMC for $q=1$, 2, 6, 8, and 26. The branches of the curve at low scaled energies correspond to $q=1$ (lower branch) and $q=6,8,26$ (upper branch). Error bars indicate statistical uncertainties. — — —, present SE for $q=1$; —·—·—·, present SE for $q=26$; · · · ·, Lodge's formula for $q=1$; △, □, ▽, ◇, ×, and ○, present AO for $q=1$, 2, 6, 8, 14, and 26, respectively.

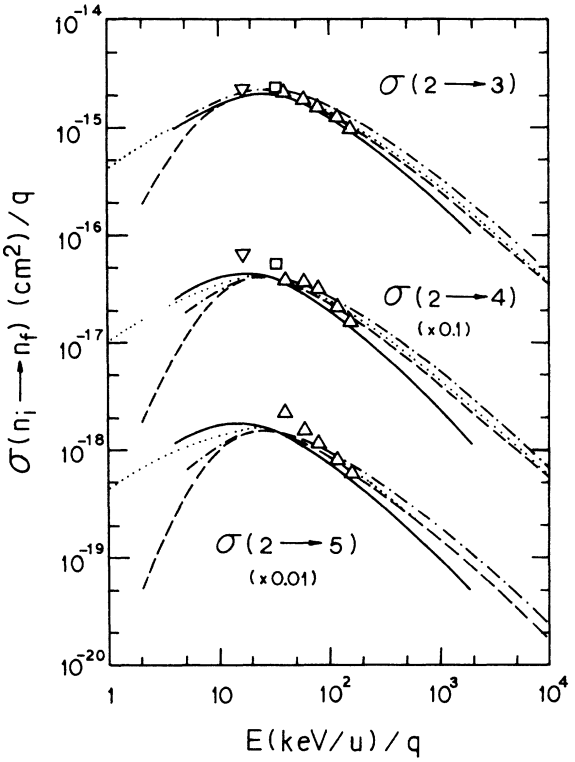

FIG. 4. Scaled total excitation cross section in $A^{q+}+\mathrm{H}(n_i=2) \rightarrow A^{q+}+\mathrm{H}(n_f=3,4,5)$ collisions as a function of the scaled impact energy. The same notation as in Fig. 3 is used.

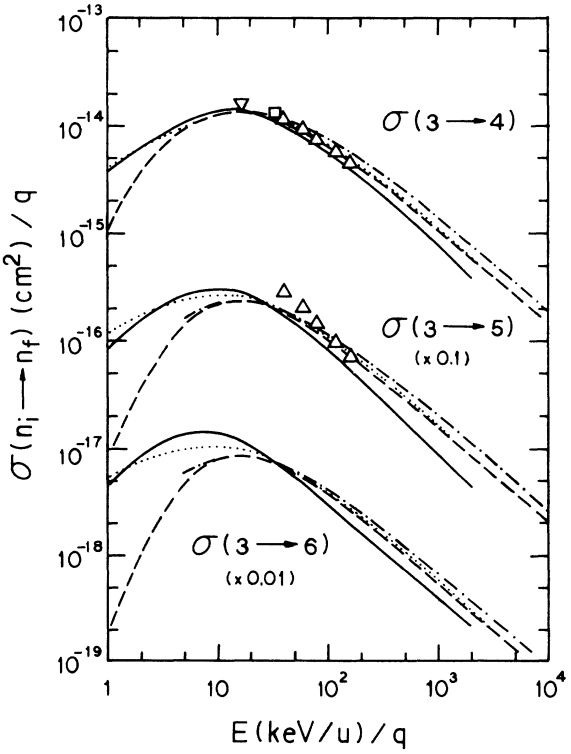

FIG. 5. Scaled total excitation cross section in $A^{q+}+\mathrm{H}(n_i=3) \rightarrow A^{q+}+\mathrm{H}(n_f=4,5,6)$ collisions as a function of the scaled impact energy. The same notation as in Fig. 3 is used.

over all the scaled energy ranges of the figures. In fact, the departures predicted by the AO and SE models for scaled energies 80 (keV/u)$/n_i^2 < E/q < 300$ (keV/u)$/n_i^2$ are very similar.

On the other hand, the departures predicted by SE at high scaled energies are real and are also predicted by the first Born approximation (i.e., the high-energy limit of SE). The reason why CTMC does not predict any departure from the scaling relation (5) at high energies is that the high-energy behavior of the CTMC excitation cross sections is proportional to q^2/E, which satisfies the scaling exactly. However, as discussed in Sec. II A, the true high-energy limit contains an additional logarithmic term that causes departures from the scaling. The largest high-energy deparatures (i.e., $E/q > 300$ keV/u) are observed for excitation from the initial level $n_i = 1$, where the scaled cross sections for Fe^{26+} projectiles differ in up to a factor of 1.55 from the cross section for proton impact. For excitation from $n_i = 2$ and 3, the departures are smaller than a factor 1.35.

Finally, we have also plotted in Figs. 1, 2, and 3 the cross sections that are obtained by means of the semiempirical formula deduced by Lodge, Percival, and Richards[29] for excitation cross sections for proton projectiles. Surprisingly, this simple formula gives cross sections in very good agreement with the best of our cross sections. That is, it agrees with our SE and AO predictions at intermediate to high scaled energies and with our CTMC predictions at intermediate-to-low scaled energies. On the contrary, we have verified (not plotted) that large disagreements exist between our calculations and the universal curves predicted by Janev and Presnyakov.[4]

Summarizing, we have demonstrated the validity of the approximate scaling relation (5) for excitation cross sections in $A^{q+}+\mathrm{H}(n_i)$ collisions by means of three independent theoretical approaches. If this scaling is combined with the semiempirical formula by Lodge, Percival, and Richards,[29] excitation cross sections in $A^{q+}+\mathrm{H}$ collisions can be written approximately as

$$\sigma(E,q,n_i \rightarrow n_f) = q\sigma^L(E/q,n_i,n_f) \tag{6}$$

where $\sigma^L(E/q,n_i,n_f)$ denotes the semiempirical cross section of Lodge, Percival, and Richards[29] for proton projectiles with impact energy E/q. Departures from (6) are expected to be smaller than a factor of 1.5 for scaled impact energies $E/q > 50$ (keV/u)$/n_i^2$. Below this impact energy range, larger departures may be expected but the present theoretical models do not allow to obtain definite conclusions about their magnitude. In fact, further theoretical and/or experimental studies are needed to determine the true magnitude of the cross sections, especially for multiply charged ions.

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