

Close-coupling study of electron excitation in 1–300-keV/u He^{2+} -H collisions

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Electron excitation to H $n=2$ and $n=3$ states in 1–300-keV/u He^{2+} -H collisions is studied within the semiclassical close-coupling method with atomic-orbital basis sets. Calculated excitation cross sections tie in smoothly with results from high-energy theories above 100 keV/u; they also agree with recently published H $n=3$ to $n=2$ line-emission cross sections [A. Donnelly, J. Geddes, and H. B. Gilbody, *J. Phys. B* **24**, 165 (1991)]. An interesting aspect of this study is that low-energy-excitation cross sections show structures that may be explained by the complex excitation mechanism. These structures are confirmed in as yet unpublished H $2p$ - $1s$ line-emission cross sections. No other information exists for energies below 10 keV/u.

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I. INTRODUCTION

In the past decade, our understanding and description of atomic collisions have much matured [1,2]. At low-to-intermediate energies, where electron transfer is the leading process, the semiclassical close-coupling description of atomic collisions has been successfully invoked in studies of total transfer and also of the more model-sensitive partial-transfer cross sections. The reliability and accuracy of the close-coupling description has been demonstrated, in particular for weak processes in one-electron systems. There are also examples of weak, two-electron transitions in two-electron collision systems that have been successfully described theoretically in spite of severe restrictions in the number and form of the basis states that can be employed in practical calculations [1,2].

Another weak process in atomic collisions is the process of target electron excitation in slow collisions. This has only rarely been considered. Much information is available on electron excitation in symmetric H^+ -H collisions where, at low velocities, the mechanisms for electron excitation and transfer become very similar. Some excitation processes have been studied, mainly experimentally, in collisions with helium targets (for a recent example, see the work by Reymann *et al.* [3]). Other studies have considered excitation processes for a few specific target systems, e.g., for alkali-metal targets where the energy defect for target excitation is small.

Virtually nothing, however, is known from experiment, and only a little more from theory, about low-energy target excitation in collisions between a highly charged ion and hydrogen atoms. Such processes, though weak in comparison with electron transfer, are important in fusion plasmas, notably for the method of plasma diagnostics by an active atomic-hydrogen beam. The prediction of electron excitation in these systems is a theoretical

challenge because it requires a high-precision description of the electron dynamics in a multilevel situation in close collisions. In this paper we present an investigation of electron excitation to the hydrogen states with principal quantum numbers $n=2$ and $n=3$ in He^{2+} -H($1s$) collisions. We will demonstrate the problems particular for this class of systems. We will further show that earlier, simpler approaches [4,5] to these systems are not satisfactory.

In the next section, we formulate the framework of our investigation. The close-coupling scheme used here will be contrasted to earlier, simpler, close-coupling methods. In Sec. III results of this work will be discussed and compared to the existing limited data base, including very recently published Balmer- α emission cross sections from Donnelly, Geddes, and Gilbody [6]. Section IV contains a few concluding remarks.

II. THEORY

For almost a decade, the pioneering work [4] by Janev and Presnyakov has provided the only source for assessing np excitation cross sections in slow collisions between highly charged ions A^{Z+} and hydrogen atoms. These authors start from the set of three coupled equations for the population of the initial $1s$ H state and the final $np(m=0)$ and $np(|m|=1)$ H states. With a number of further simplifying assumptions they arrive at an analytic expression for excitation cross sections which are found to lie, after suitable scaling, on a universal curve for all charge states Z .

As has been pointed out [5], the simplifying assumptions used by Janev and Presnyakov are rather severe. A mere three-state description in the spirit of Janev and Presnyakov but without any of their further approximations has been shown [5] to lead to substantially different excitation cross sections at low energies. Indeed, the results from a *consistent* three-state model [5] for $2p$ excita-

tion do not display the Janev-Presnyakov scaling except in the Born regime at high energies.

Clearly, a three-state model for the description of low-energy electron excitation is not satisfactory. Fritsch and Schartner have also used [5] much larger basis sets of atomic states to study $2p$ excitation for a few specific systems. These basis sets consist of a number of excited atomic target states and ionization pseudostates, as well as the dominant transfer states of the projectile. The results from such improved calculations were seen to approximately scale for energies above some 15 keV/u Z except for proton projectiles ($Z=1$), the approximate universal curve being different in slope and magnitude from the curve given by Janev and Presnyakov. Thus it is clear that a multistate close-coupling study is needed for a reliable prediction of low-energy excitation cross sections. It is less clear from the outset whether excitation cross sections from the schematic procedure used by Fritsch and Schartner are quantitatively reliable.

In this work we strive for an improved description of electron excitation in the He^{2+} -H collision system. Figure 1 shows the diagram of molecular energies [7] for this system. In slow collisions, the electron may proceed from its initial $1s$ H ($2p\sigma$) state to the set of final $2s, 2p(5g\sigma, 4f\pi, 4d\sigma)$ H states by a sequence of steps: (1) the radial $2p\sigma$ - $3d\sigma$ coupling may effect the promotion of the electron to the band of $n=3$ united-atom orbitals; (2) further couplings there, notably the $3d\sigma$ - $3d\pi$ - $3d\delta$ rotational coupling at small separations, may promote the electron to higher energies, most of them ending up from this point in the $n=3$ He^+ capture states; (3) finally, couplings to the final $5g\sigma, 4f\pi$, and $4d\sigma$ states may lead to the small population of excitation states; of these, the $3d\pi$ - $4f\pi$ radial coupling may be particularly effective, leading to the predominant population of $2p(|m|=1)$

states of H within the $n=2$ states. Other paths to the population of $n=2$ H states are also possible, such as the sequence of $3d\sigma$ - $4f\sigma$ - $5g\sigma$ radial couplings; this latter path is expected to gain strength with increasing velocities. The role of processes (1) and (2) for electron transfer in He^{2+} -H collisions has been discussed in work within the molecular-orbital expansion framework [8-11]. The population of excited H $n=3$ states is even less favorable in slow collisions. It requires a number of additional steps, or else the direct interaction between non-neighboring states.

A reliable description of low-energy electron excitation in this system hence requires the inclusion or representation of a number of molecular orbitals, including those with high angular momentum numbers in the united-atom limit, and those correlating, in the separated-atom limit, to the states of the sub dominant capture channel $\text{He}^+(n=3)$ - H^+ . We have therefore studied He^{2+} -H collisions within the semiclassical close-coupling method, using a set of two-center atomic basis states that includes united-atom orbitals as pseudostates [12] at the two centers (AO+basis). The choice of states is guided by consideration of the important molecular orbitals as discussed above: the states should represent the important molecular states at both infinite and vanishing interatomic separations exactly, they should then represent the molecular orbitals approximately at finite separations. Table I shows the hydrogenic states included in this basis, 31 at the H center and 23 at the He center. All states deemed to be important for hydrogen $2s$ - and $2p$ -excitation slow collisions are included in the basis. The choice of center for the pseudostates is somewhat ambiguous in the AO+ description. We have taken the H center for the $4d, 4f$, and $5g$ ($Z=3$) states which should help the representation of the molecular orbitals correlating to the H $n=2$ states. The $n=2$ ($Z=3$), $4f$ ($Z=3$), and $3l$ ($Z=3$) states have been included at both centers as they presumably play the key role in promoting the electron to the excited states. We note that the use of

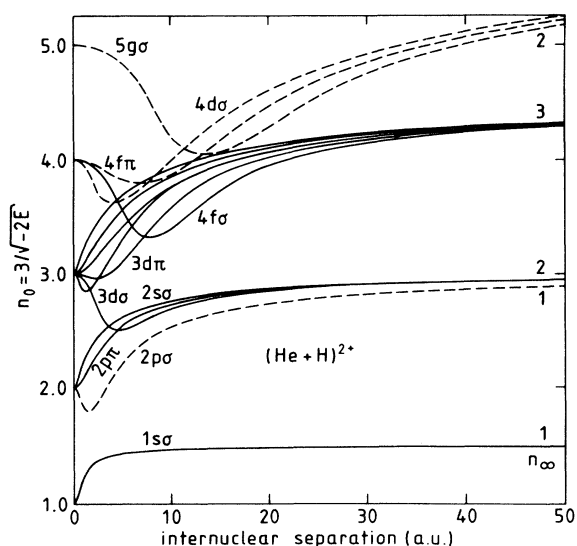


FIG. 1. Molecular energies [7] of the $(\text{He} - \text{H})^{2+}$ collision system. Solid lines correlate to states in He^+ , broken lines to states in H.

TABLE I. AO+ basis set for the description of low-energy excitation in He^{2+} -H collisions. Given are for each state at a given center the designation of hydrogenic orbitals: the principal quantum number n , the angular momentum quantum number (the entry l means the full set of l quantum numbers for a given n), and the charge number Z . For each combination (n, l, Z) , the full set of m quantum number is included in the basis.

Center H			Center He		
n	l	Z	n	l	Z
1	s	1.0	1	s	2.0
2	l	1.0	2	l	2.0
3	l	1.0	3	l	2.0
2	l	3.0	2	l	3.0
3	l	3.0	3	l	3.0
4	d	3.0	4	f	3.0
4	f	3.0			
5	g	3.0			

equivalent states at the two centers does not introduce any problem: states at different centers move towards each other with the collisional speed of those centers, the associated plane-wave translation factors $\exp(\pm i\mathbf{v}\cdot\mathbf{r})$ of otherwise identical states are different; even at vanishing interatomic separations the basis is still not linearly dependent (moreover, vanishing interatomic separations do not occur in real collisions and also not in the determination of cross sections). The details of the calculations with the AO+ basis set are the same as in earlier work [12,2]. We note that H $n=3$ states are included in the AO+ basis, but pseudostates which may help the representation of associated molecular states are not.

In work with AO+ basis sets, deficiencies of pure AO descriptions of atomic collisions are addressed by a choice of pseudostates [2,12] which allows a close representation of molecular orbitals in close or slow collisions. In this work, we have studied electron excitation in He^{2+} -H collisions also with a second choice of pseudostates in the atomic basis, here for distinction called an AO-C basis. Also this AO-C basis is chosen to include the dominant atomic bound states of the colliding partners. These states are further augmented with some bound and positive-energy pseudostates placed on the respective atomic centers. The positive-energy pseudostates represent the ionization channels populated in the collision at intermediate and high impact velocities. In this respect the basis states are designed to satisfy the oscillator-strength sum rules. Furthermore, these positive-energy pseudostates represent the corresponding Coulomb wave function in the important region of space of bound states' dimensions. Experience has shown [13] that such basis set is also a prudent choice at low impact energies.

Since we are interested in target excitation to H $n=2$ states in the He^{2+} -H collision system, the exact $n=1$ to 3 states of the hydrogen atom and the exact $n=1$ to 4 states of the singly ionized helium ion were included in the AO-C basis set. It may be noted that the $n=4$ state of the He^+ ion is resonant with the $n=2$ hydrogen-atom state. The eigenstates were obtained by diagonalizing the one-electron Hamiltonian in a basis of Slater-type orbitals. The target (H) was represented by 22 atomic states described in Table I of Shingal, Bransden and Flower [14]. The He^+ atom was described in terms of 38 atomic states, 10 s -type orbitals, 6 p -, 4 d - and one f -type orbital. The parameters and the energy expectation values are displayed in Table II. It is noted that exact $n=1$ to 3 states for the hydrogen atom and exact $n=1$ to 4 states of the singly ionized helium ion are included through this basis set.

The two basis sets used in this work differ mostly in the choice of pseudostates, except for the atomic $\text{He}^+n=4$ states which are included in the AO-C basis but not in the AO+ basis. In the AO+ basis, there are more states with high angular momentum quantum number l , in the AO-C basis there are more pseudostates with small decay constant λ . Of course it is hoped that the main physics of the collision is well described with *both* sets of basis states in some range of collision energies, and actually the results show that this is the case. On the other hand, pseu-

TABLE II. AO-C basis set for the description of excitation in He^{2+} -H collisions at higher energies. Given are the parameters of Slater-type orbitals $r^{p+1}\exp(-\lambda r)Y_l^m(\theta\phi)$ at the He center. Also given are the energies ϵ from diagonalizing the He^+ Hamiltonian in the set of basis states. The basis states at the H center are given in Table I of Shingal, Bransden, and Flower [14].

l	p	λ	$\epsilon(\text{au})$
0	0	2.0000	-2.0000
	0	1.0000	-0.5000
	1	1.0000	-0.2222
	0	0.6667	-0.1250
	1	0.6667	-0.0798
	2	0.6667	-0.0431
	0	0.5000	0.0413
	1	0.5000	0.2849
	2	0.5000	1.1844
	3	0.5000	7.5708
1	0	1.0000	-0.5000
	0	0.6667	-0.2222
	1	0.6667	-0.1250
	0	0.5000	-0.0758
	1	0.5000	0.0313
	2	0.5000	0.7150
2	0	0.6667	-0.2222
	0	0.5000	-0.1250
	1	0.5000	-0.0545
	0	1.2753	0.4381
3	0	0.5000	-0.1250

dostates cannot be chosen deliberately. We have tried various smaller basis sets in test calculations and found that cross sections for the weak H 2s-excitation channel depend very sensitively on the presence and the specific choice of pseudostates. More remarks on other choices of basis sets are included in the discussion of the next section.

III. RESULTS AND DISCUSSIONS

Since there is only little known about excitation processes in this collision system, and since the excitation process is of sufficient complexity, the reliability of calculated excitation cross sections is of some concern. As it turned out, excitation cross sections for H $2p$ and $2s$ states, calculated with the two sets of basis sets, agree closely over a range of energies. Figure 2 shows the impact-parameter dependence of calculated probabilities for excitation to H $2p$ and $2s$ states at 10 keV/u impact energy. The results from the two basis expansions are seen to agree very well for the $2p$ -excitation channel and still well for the weaker $2s$ -excitation channel. We note that this agreement is nontrivial in a situation when the excitation channels are populated with small probabilities, and when the competing transfer channels are much stronger. Figure 2 includes the probabilities for transfer into the $\text{He}^+n=3$ states; the probabilities for the $\text{He}^+n=2$ states (not shown) are about an order of magnitude larger. The $3l$ -excitation cross sections from the two

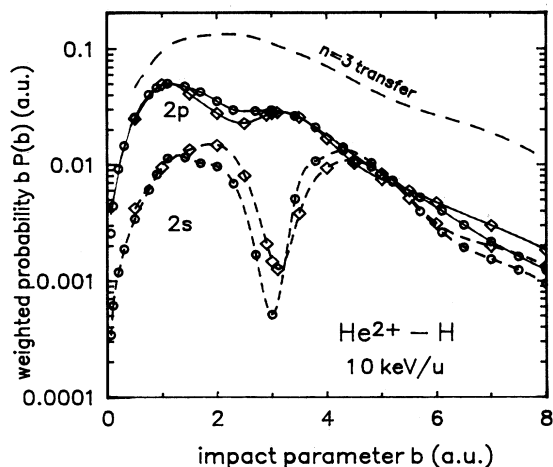


FIG. 2. Calculated weighted transition probabilities $bP(b)$, in a range of impact parameters b , for H $2s$, H $2p$, and the set of $\text{He}^+ n=3$ states, in 10 keV/u He^{2+} -H collisions. Results shown as diamonds are calculated with the AO+ basis, circles are calculated with the AO-C basis.

calculations, on the other hand, are even smaller than the $2s$ -excitation cross section. The two calculations agree only to within a factor of 2.

Calculated excitation cross sections are listed in Table III, a subset of results is displayed in Fig. 3. In both cases, results at 10 keV/u and above are taken from the AO-C calculation, results below that energy from the AO+ calculation. The excitation cross sections to H $n=2$ states and to the $2s$ substate show pronounced structures, and so to a lesser extent do the cross sections for excitation to the set of H $n=3$ states. From the agreement between calculations with two different basis sets, we believe that these structures are real, certainly for the $2s$ - and $2p$ -excitation channels. Also included in Fig. 3 are the calculated [15] cross sections for ionization in He^{2+} -H collisions. These cross sections increase faster with energy than do the excitation cross sections and become larger than the latter around 10 keV/u . Clearly, at such high energies, interactions between excitation and ionization channels may be important; these interactions are taken into account in basis set AO-C, to the extent that the actual continuum is represented by the pseudo-states in the basis.

In Fig. 4 we compare the results calculated in this work with other information available for this collision system. At high energies, the calculated H $2p$ -excitation cross sections of this work are seen to tie in smoothly with results from other sources: the semi empirical formula due to Lodge, Percival, and Richards [16] that gives an assessment of excitation cross sections for the H^+ -H system and which may be scaled [4] to the class of Z^{Z^+} -H systems (such scaling is customary in applications); the results from the classical-trajectory Monte-Carlo calculations (CTMC) and from the symmetric eikonal approach (SE) by Reinhold, Olson, and Fritsch [17]; the result from the three-state model study by Janev and Presnyakov [4]; and the results from the many-state study by Fritsch and

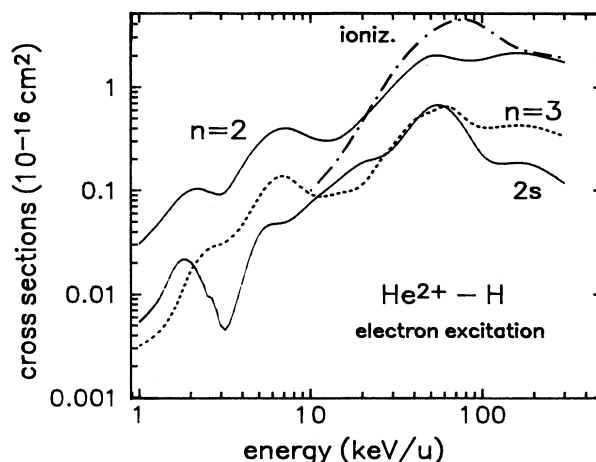


FIG. 3. Calculated excitation cross sections in He^{2+} -H collisions to H $2s$ states (lower solid line), to the set of H $n=2$ states (upper solid line), and to the set of H ($n=3$) states (dotted line), all from this work. Calculated ionization cross sections (dash-dotted line) are from Ref. [15].

Schartner [5]. At energies well beyond 100 keV/u , all these curves should become close to the excitation cross section from Born-type descriptions. Around 100 keV/u , however, the curve from this work already shows structure that make it distinct from the smooth curves of the high-energy approximations SE and CTMC. Also deviations from the scaled curve by Lodge, Percival, and Richards become appreciable. Around 100 keV/u , the deviation between the excitation cross sections from this work and the curve by Janev and Presnyakov shows that excitation does not occur, at these intermediate energies, by the exclusive interplay of the excited states with the initial state. Deviations from the curve by Fritsch and Schartner indicate that careful account of the electron continuum is needed, as has been tried in this work.

At still lower energies, around 10 keV/u and below, the excitation curve due to Lodge, Percival, and Richards is still a rough representation of the curve from this work but the details of the latter are missing. The deviation of excitation cross sections from this work and from the earlier close-coupling study by Fritsch and Schartner shows that the particular excitation mechanism needs careful consideration at low energies. Unpublished line-emission cross sections by Hoekstra and Beijers [18] (no cascade corrections applied) have been identified as $2p$ -excitation cross sections. They turn out to confirm the calculated structure around 7 keV .

The excitation cross sections to H $n=3$ states may be expected to be less reliable than the stronger cross sections to H $n=2$ states. In a restricted energy regime, they can be tested by comparing Balmer α emission cross sections from the recent experiment by Donnelly, Geddes, and Gilbody [6] and from the calculations. As shown in Fig. 4, the two sets of results agree closely, actually more closely than one would expect for such a weak channel. No data is available in the energy region where the calculations predict structure.

We have tried to identify the cause for the pronounced structure of calculated excitation cross sections. For such analysis it would be necessary to isolate, in actual calculations, the various excitation mechanisms and to study their interplay. While the large pseudostate expansions of this work are not well suited for use in qualitative discussions, it turned out that selected smaller expansions lead to excitation cross sections that deviate strongly from the results in Table III. (Smaller basis sets can be devised for calculations which are still aimed at predicting reliable H $2p$ cross sections but not H $2s$ cross sections. Such basis sets are, however, still too large for use in a qualitative discussion of excitation mechanisms.) Moreover, the overlap of atomic states at small interatomic separations tends to render an interpretation of the physical content of a given basis ambiguous in close collisions. Hence, in the absence of a molecular study for excitation channels, the following qualitative arguments may suffice.

From studies [10,11,19–23] of electron transfer in He^{2+} -H collisions (see also references cited in Refs. [1] and [2]) it is known that the cross sections for transfer into He^+2s and $2p$ states display a broad peak around an energy of 10 keV/u. At about this energy, the cross section for transfer into $\text{He}^+n=3$ states displays [23] a shoulder in its rise with energy, the maximum being at

about 30 keV/u. It appears, therefore, that the $2p\sigma$ - $3d\sigma$ radial coupling (main mechanism for transfer into $\text{He}^+n=2$ states) is most efficient around 10 keV/u. On the other hand, the mechanism for subsequent population of the $3d\pi$, $3d\delta$, $3p\sigma$, $3p\pi$, and $4f\sigma$ states, a mixture of rotational couplings and delocalized radial couplings, is most efficient at the higher energies around 30 keV/u, where it might also be termed “direct population” (note that the assessment of the $n=3$ transfer channel within the unitarized distorted-wave approximation (UDWA) [24] predicts already the main portion of the cross section as calculated in the close-coupling scheme [23], around its maximum). Low-energy electron excitation to H $n=2$ states proceeds from the latter states to the $4d\sigma$, $4f\pi$, and $5g\sigma$ states [The $3d\pi$ - $4f\pi$ radial coupling may be considered to contribute prominently to the excitation mechanism since, according to the calculations of this work, H excitation is mostly to the H $2p$ ($|m|=1$) state.] Such couplings are not localized, hence turns of the phases in each coupling and, more importantly, the contributions of various couplings to the population of a given excited state may then well lead to strong structures in the impact-parameter dependence of transition probabilities and weaker structures in the energy dependences of cross sections. A more detailed understanding, particularly of the structures of cross sections at low en-

TABLE III. Calculated cross sections (in 10^{-16} cm²) for excitation of hydrogen atoms in He^{2+} -H(1s) collisions.

E (keV/amu)	$2s$	$2p$	$3s$	$3p$	$3d$
1.00	0.0054	0.025	0.0002	0.0012	0.0017
1.40	0.0112	0.045	0.0006	0.0012	0.0032
1.70	0.0206	0.062	0.0010	0.0031	0.0049
2.00	0.2022	0.081	0.0027	0.0068	0.0075
2.20	0.0162	0.088	0.0038	0.0092	0.0087
2.50	0.0096	0.088	0.0054	0.0111	0.0104
2.70	0.0067	0.085	0.0056	0.0112	0.0121
3.00	0.0049	0.086	0.0047	0.0104	0.0153
3.50	0.0061	0.111	0.0046	0.0093	0.0220
4.10	0.0148	0.167	0.0037	0.0156	0.0310
5.00	0.0371	0.248	0.0057	0.0425	0.0394
7.00	0.0488	0.354	0.011	0.089	0.038
10.00	0.0757	0.251	0.013	0.036	0.035
15.00	0.1310	0.197	0.031	0.036	0.028
20.00	0.1860	0.309	0.043	0.044	0.027
25.00	0.2080	0.517	0.046	0.097	0.046
30.00	0.2530	0.763	0.049	0.151	0.075
40.00	0.4550	1.161	0.094	0.258	0.129
45.00	0.5670	1.295	0.114	0.281	0.144
50.00	0.6490	1.354	0.137	0.284	0.157
55.00	0.6770	1.352	0.155	0.309	0.165
60.00	0.6520	1.328	0.178	0.312	0.163
70.00	0.5220	1.336	0.174	0.273	0.149
75.00	0.4510	1.368	0.140	0.258	0.142
90.00	0.2830	1.522	0.097	0.206	0.127
100.00	0.2220	1.635	0.053	0.234	0.119
125.00	0.1810	1.859	0.031	0.278	0.107
150.00	0.1850	1.954	0.031	0.301	0.096
200.00	0.1740	1.880	0.032	0.309	0.073
300.00	0.1170	1.622	0.012	0.277	0.044

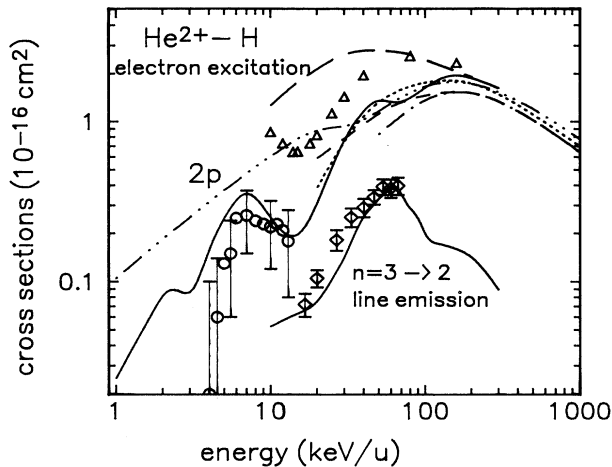


FIG. 4. Calculated excitation cross sections to H $2p$ states (upper solid line) and calculated H $n=3$ to $n=2$ line-emission cross sections (lower solid line), all from this work, are compared to results for H $2p$ excitation cross sections from the three-state model by Janev and Presnyakov [4] (long-dashed line), from the many-state (mainly target centered) study by Fritsch and Scharner [5] (triangles), from the CTMC work in Ref. [17] (short-dashed line: scaled results derived with projectiles $Z \geq 6$, dash-dotted line: scaled results derived with $Z=1$), from the symmetric eikonal approach in Ref. [17] (dotted line), from the semiempirical formula due to Lodge, Percival, and Richards [16] (dash-triple-dotted line), and to measured $2p-1s$ line-emission cross section [18]. Experimental H $n=3$ to $n=2$ line-emission cross sections are by Donnelly, Geddes, and Gilbody [6] (diamonds).

ergy, has to wait a study within the molecular model of atomic collisions.

IV. CONCLUSIONS

In this work we present a study of low-energy electron excitation in He^{2+} -H collisions, extending from 1 up to 300 keV/u. The calculated H $2p$ -excitation cross sections tie in smoothly with what is available from high-energy theories. At low energies, these cross sections are

confirmed by unpublished line-emission cross sections [18], they deviate appreciably from results which are generated by scaling, in a questionable manner, excitation cross sections from the H^+ -H system. We believe that the H $n=2$ excitation cross sections of this work constitute reliable predictions within about 20%. The calculated H $n=3$ excitation cross sections that are expected to be less reliable, still agree well with measured $n=3$ to $n=2$ line-emission cross sections above 15 keV/u.

Perhaps most strikingly, in the calculated excitation cross sections we note a number of structures which we attribute to the complex mechanism of populating target excited states. We discuss, on the basis of the static molecular-energy diagram, the various paths that lead to the population of excited hydrogen states. For a full analysis, a dynamical study within the molecular model of atomic collisions would be desirable. Obviously, experimental confirmation of the structures are also needed.

This study shows that rather large basis sets are needed for predicting excitation cross sections in low-energy He^{2+} -H collisions. In the present case, it appears that low-energy excitation proceeds through three series of couplings. Of course, the density of states in the molecular-energy diagram has its correspondence in the density of important atomic states in a study of excitation at higher energies. It is only in the Born regime that electron excitation becomes a simple, one-step process again.

Clearly, the situation is even more complex when it comes to describing electron excitation in higher charged collision systems. Questions which might be addressed are the following: Will oscillatory structures appear also in higher charged systems or will they be washed out due to the multitude of possible excitation paths? Does approximate scaling of excitation cross sections, within a factor of about 2, still prevail? It is hoped that this study helps in arousing interest in such systems from both the experimental and the theory points of view.

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