

Atomic and Molecular Databases for Fusion Divertor Plasmas

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Abstract. We describe our recent activities regarding the production of atomic and molecular data that are needed for modeling divertor plasmas in fusion tokamaks. The transport of particles and, in particular, the exchange of momentum in such plasmas can be dominantly influenced by elastic scattering and resonant charge transfer among hydrogen ions, atoms, and molecules. We have undertaken a comprehensive calculation of these processes, in all isotopic combinations of hydrogen, and highlights of that study, with intercomparison of various integral and differential cross sections, are shown here. Also, in the formation of the detached plasma layers the processes of charge transfer between hydrogen ions and vibrationally excited molecules might play a crucial role in the chain of reactions called Molecule Assisted Recombination (MAR). All other inelastic processes involving hydrogen molecules are also of interest in the colder parts of the divertor plasma. We have studied all processes that involve hydrogen ions/atoms and vibrationally excited molecules/molecular ions (charge transfer, excitation, dissociation, association), in the range of Center-of-Mass (CM) collision energies 0.5-10 eV. Here we discuss integral cross sections for these reactions, resolved in both initial and final vibrational states. The full set of the cross sections for both elastic and inelastic processes which involve molecules can be obtained through the ORNL Controlled Fusion Atomic Data Center's website.

1. INTRODUCTION

The Oak Ridge national Laboratory (ORNL) Controlled Fusion Atomic Data Center (CFADC) has multiple goals defined by the needs of the fusion community, which can be organized into several categories: the production of atomic collision data, compilation of numerical and bibliographic data, dissemination of the data to the fusion community, and theoretical support for ORNL atomic collision experiments. These goals are addressed by the development of new theoretical methods or utilization of existing ones as well as by compilation of bibliographic data by a network of consultants. Besides the need for improved understanding of the relevant atomic processes, the production of data is characterized by the need for comprehensive calculations of the cross sections over a sufficiently wide energy range to determine the rate coefficients. The data are disseminated by publication in scientific journals and on the CFADC [www site \(www-cfadc.phy.ornl.gov\)](http://www-cfadc.phy.ornl.gov).

It is widely recognized in the fusion community that the divertor region is of crucial importance for the successful engineering, design, and operation of tokamak fusion reactors, while the atomic collision data needed to model and diagnose this region are often unknown or only poorly considered in the extant literature. The high density, low temperature divertor plasma is characterized by similar densities of ions and neutrals, consisting predominantly of hydrogen atoms, ions and molecules in various isotopic combinations. The transport of particles and, in particular, the exchange of momentum in this plasma can be dominantly influenced by elastic scattering and resonant charge transfer among hydrogen ions, atoms, and molecules. We have recently undertaken a very large set of fully quantal calculations to remedy this need. Four types of systems, ion-atom (P^+Q), atom-atom ($P+Q$), ion-molecule (P^+QR) and atom-molecule ($P+QR$) have been studied [1-4], where P, Q, and R are any of the hydrogen isotopes (H, D, or T). In addition, P^+He [1,2], P^+C [5] and P^+Ar systems were studied, as well. Several thousands of accurate differential and integral cross sections were calculated, spanning the center of mass (CM) collision energy range of 0.1-100 eV.

In addition, formation of the divertor plasma detachment layer might critically depend on Molecule Assisted Recombination (MAR), which involves ion conversion via electron capture by a proton from a vibrationally and rotationally excited hydrogen molecule, followed by dissociative recombination of the molecular ion with plasma electrons. Such collisions require accurate data for the relevant processes, particularly for vibrationally resolved charge transfer from H_2 to H^+ . We report on a comprehensive study of scattering of hydrogen ions on vibrationally excited hydrogen molecules as well as of hydrogen atoms on vibrationally excited hydrogen molecular ions in the range of center of mass energies 0.5 - 10 eV. Total and partial, initial and final vibrational state resolved cross sections for excitation, charge transfer, dissociation (including dissociative energy spectra), and association have been calculated “on the same footing” using a fully-quantal, coupled-channel approach. An extensive vibrational basis set of several hundred states, including discretized dissociative continua in a large configuration space (to include nuclear particle arrangements) was employed, while the rotational dynamics of H_2 and H_2^+ were treated within the sudden approximation. Concerning the details of the treatment of the reactive dynamics, the data summarized here are an improvement over these previously published [6]. Still, limitations of the sudden approximation require extension of the method to fully include rotational dynamics if isotopic variations of hydrogen are employed. These will be the subject of future research.

In Section 2 we discuss the elastic processes. We describe our study of inelastic processes involving molecules and molecular ions in Section 3.

2. ELASTIC AND TRANSPORT COLLISION PROCESSES IN A COLD HYDROGEN PLASMA

Elastic scattering among ions, atoms, and molecules can cause dissipation of the divertor plasma momentum. Neutrals, produced through recombination processes, can carry off a part of the collision energy, but are not controlled by the magnetic field and

thus diffuse to the walls unless re-ionized. In the detached plasma regime diffusion of neutrals is crucial.

We have performed a study of elastic processes for the $H^+ + H$, $H + H$, $H + H_2$, and $H + H_2^+$ systems, in all combinations of the hydrogen isotopes for both projectiles and targets, in the CM collision energy range 0.1-100 eV, with ten energy values per decade. In addition, the systems $H^+ + He$, $H^+ + C$, and $H^+ + Ar$ were studied for all isotopic combinations of H. The calculation resulted in over 3000 differential and more than 250 integral cross sections, describing elastic scattering (EL), and transport of momentum (momentum transfer, MT) and energy (viscosity, VI), charge transfer (CT), and spin exchange (SE). These can all be found at the CFADC www site (www-cfadc.phy.ornl.gov).

Details of the study can be found in recent publications [1-5] so here we summarize only the most important points.

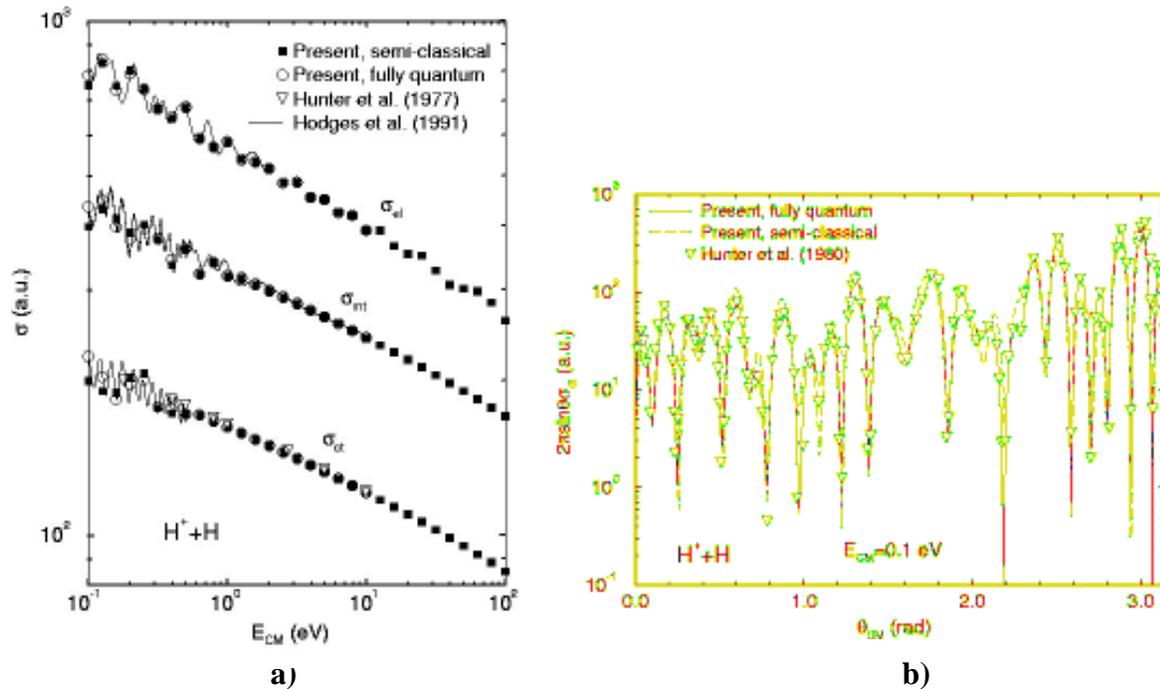


FIGURE 1. a) $H^+ + H$ integral elastic, momentum, and charge-transfer cross sections, compared with calculations of Hunter et al. (1977) [7] and Hodges et al. (1991) [8]; b) Differential cross section $H^+ + H$ at low energies compared with the results of Hunter et al. (1980) [9].

In studying symmetric ion-atom and atom-atom collisions ($H^+ + H$, $D^+ + D$, $T^+ + T$, as well as $H + H$, $D + D$ and $T + T$) an important problem arises from the indistinguishability of the projectile and the target particles [1-3]: in the ion-atom cases one cannot distinguish at the exit channel the elastically scattered projectile (A) from the nucleus coming from charge transfer (B) unless these particles are somehow labeled. While classical mechanics allows such labeling, quantum mechanical wave packets are not well localized at the lowest energies, allowing for overlap and thus, interference of channels A and B. Only tracking of the nuclear spin of these particles can be used for positive identification of the elastically scattered and charge transfer channels, which is neither typical for common experiments nor plasma environments.

One rather averages over the spins in the input and sums them in the output channel which produces a quantity called the elastic cross section, although it is made of a coherent sum of elastically scattered and charge transferred ions, which we denote the IP (Indistinguishable Particle) elastic cross section in our www database. The integral moments (momentum transfer and viscosity) (Fig. 1a) calculated from the IP differential cross sections (Fig. 1b) define the transport due to both elastic scattering and charge transfer. This needs to be explicitly taken into account in particle transport modeling to avoid double counting. On the other hand, for CM collision energies above 1 eV, most of the charge transfer differential cross section can be distinguished by its strong peak in the backward direction, with weak overlap with the elastically forward scattered particles, i.e. one can label the scattered particles A and B by their different angular distributions, which allows for “standard” distinguishable particle (DP) definitions of the transport moments. For modelers whose codes require such distinguishability, we provide also “labeled

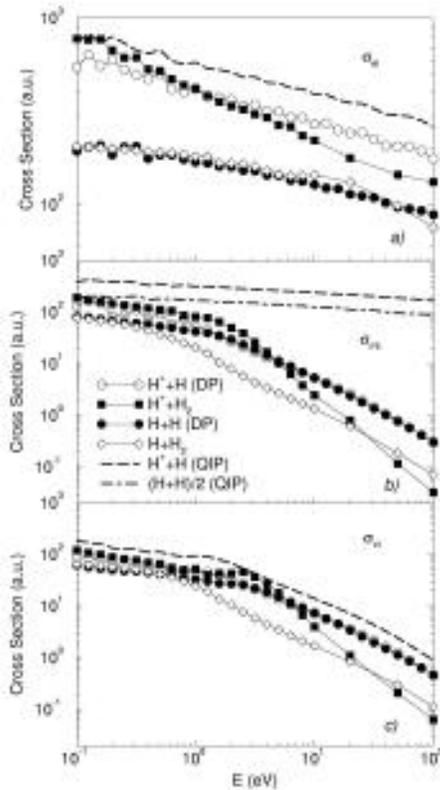


FIGURE 2. Effects of the symmetry of the colliding hydrogen ions, atoms, and molecules.

particle” elastic cross sections and moments (denoted by DP in our www database). Still, the DP cross sections introduce an error of about 10% at energies below 1 eV, due to neglect of interference effects [3].

In the case of symmetric atom-atom collisions, a similar problem emerges due to indistinguishability of the elastically scattered and recoiled particles, which disappears at higher energies due to the decreasing overlap between the elastic (forward) and recoil channel (backward angles) [1,2]. Due to the full symmetry of the problem, the integral elastic cross section follows exactly its “classical” counterpart by simple division of the total integral cross section by a factor of 2. On the other hand, we recognize that, after correct averaging-summing over the nuclear and electron spins, the elastic cross section is identical to the momentum transfer cross section in symmetric atom-atom scattering [1,2,10].

The behaviors discussed are illustrated in Fig. 2 for hydrogen. For ion/atom-molecule scattering, as well as for ion/atom-atom scattering with isotopically different particles, the problems

arising from the symmetry of the particles are not present, yielding the standard elastic and charge transfer cross sections.

Another important issue is the accuracy of the data. By choice of highly accurate intermolecular potentials as well as by controlling the accuracy of the computational procedures, 7 digit numerical accuracy was reached for ion-atom collisions [1,2]. Still, the physical accuracy was far worse, caused by neglect of inelastic processes, mainly

at the higher end of the energy range considered. Thus we found that inclusion of inelastic charge transfer processes in ion-atom collisions changed the integral cross sections by 3% for viscosity, 2% for momentum and charge transfer, and 1% for “pure” elastic scattering. For the atom-atom scattering, the contribution of neglected inelastic channels, electronic excitation of the projectile or target and dissociation, is significantly smaller in the energy range considered. Concerning the cross sections for ion/atom-molecule cases, up to nine vibrationally excited states were taken simultaneously into account in the calculation of the elastic channels, while charge transfer (of importance at higher energies) was neglected. In addition, the rotational motion of the target was frozen during the collision, and the cross sections averaged over different orientations of the molecule (Infinite Order Sudden Approximation, IOSA, [11-13]), which might produce some error at the lowest energies considered. Having in mind all these effects, the overall physical uncertainty of the integral cross sections over the whole energy range (0.1-100 eV) is estimated to be less than 1% for ion-atom and atom-atom scattering, and less than 10% for the ion/atom-molecule systems considered. In Fig. 3a we show a comparison of our differential cross sections for vibrational excitation of H₂ by proton impact (dashed line) with the experimental results of Herman et al. [14]. The solid lines in the same figure are for vibrational excitation of H₂ by impact of hydrogen.

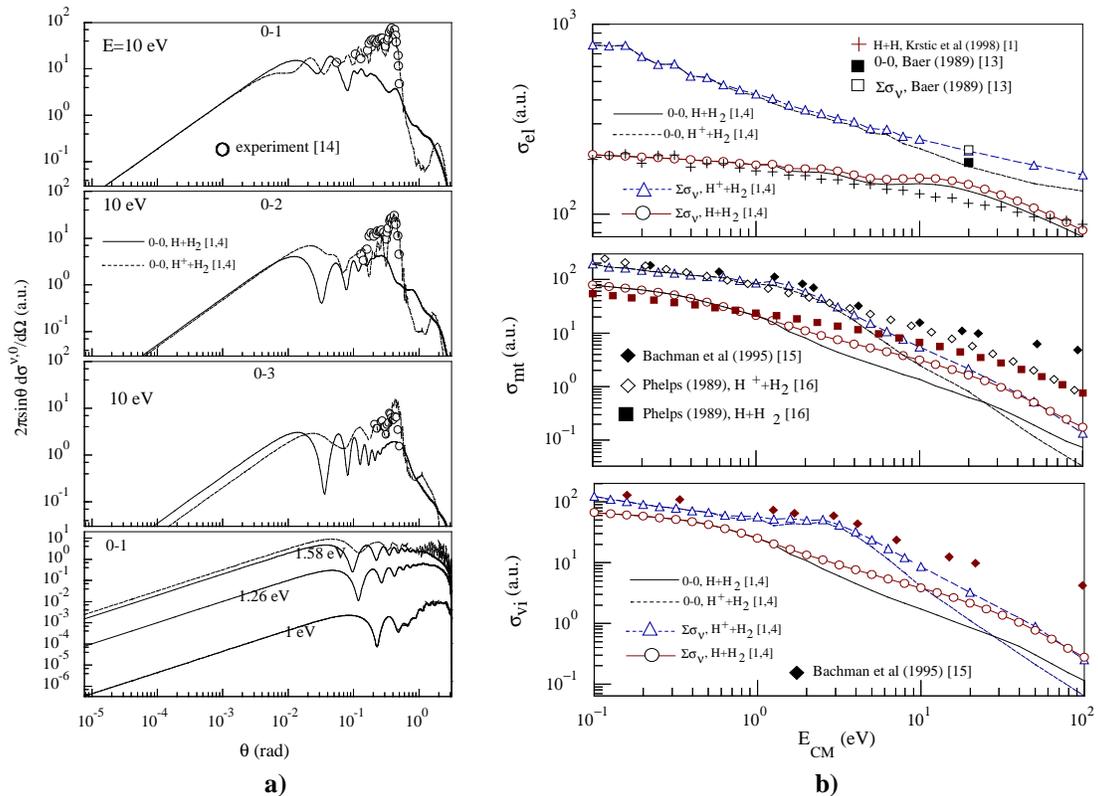


Figure 3 a) Differential elastic cross sections for vibrational excitation in H⁺, H+H₂ scattering; b) Elastic, momentum transfer, and viscosity cross sections for the ground vibrational state of H₂.

Next important issue in ion/atom-molecule scattering is the depletion of the ground vibrational state due to vibrational excitation. The effect is visible in Fig. 3b, where

the elastic, momentum transfer, and viscosity cross sections are compared with evaluated cross sections of Phelps [16]. The latter were obtained by interpolation of the measured data at about 1 eV, and calculated data at several hundred eV collision energy. In the intermediate, interpolated region of energies, where the vibrational processes are the most intensive, the deviation of Phelps' data, induced by neglecting the depletion of the ground vibrational state reaches an order of magnitude. On the other hand, the agreement of our data with that of Phelps is very good in the region of the measured data.

To summarize, we list the highlights of the database of elastic cross sections for the hydrogen collision systems:

- Benchmarks were set for both differential and integral cross sections (elastic, momentum transfer, viscosity and charge transfer) for ion-atom and atom-atom collisions involving hydrogen atoms in all isotopic combinations.
- Scaling with isotopic mass was found for the integral cross sections and their moments for all cases studied (including molecules) [1-5].
- Momentum transfer and viscosity cross sections were shown to be independent of isotopic composition, to a large extent [1-5].
- New definitions of the elastic cross sections were introduced to deal with inconsistencies in the modeler's implementations of the data in symmetric systems [3].
- Inadequate transport formulas in the literature were corrected [1,3,10].
- By inclusion of vibrational excitations, the cross sections for molecular targets were significantly improved [1,4], in the vibrationally intensive region of energies, over the previous published ones [16].
- All differential and integral cross sections were fitted to analytical expressions. These were published in Ref. [1].
- Finally, all raw data are available in both tabular and graphical form through the CFADC web site www-cfadc.phy.ornl.gov.

3. INELASTIC PROCESSES IN SLOW COLLISIONS OF IONS AND ATOMS WITH VIBRATIONALLY EXCITED MOLECULES

We have also produced a comprehensive database for inelastic, vibrationally resolved processes on the two lowest adiabatic electronic surfaces of the H_3^+ complex, from all vibrationally excited states of H_2 and H_2^+ for collisions of $H^+ + H_2$ and $H + H_2^+$, in the CM energy range 0.5-10 eV. As described in the introduction, the principal motivation for this study was to improve the very incomplete knowledge base regarding the potentially important process of MAR in diverter plasmas.

Only one comprehensive data set exists for the charge transfer from excited vibrational states of H_2 , calculated by the Trajectory Surface Hopping (TSH) method [17]. Due to the classical nature of the TSH method, this approach might not give reliable data in the eV energy range. We performed a fully quantal calculation of the vibrationally resolved processes [6,18], where electronic and vibrational dynamics are

completely taken into account, while the rotational motion of the diatom was treated within IOSA, as in Section 2. The rearrangements channels were included implicitly by doing calculations in a large configuration space of nuclear motion (encompassing 40 a.u. in all directions.) The role of dissociative continua (of both H_2 and H_2^+) was taken into account by discrete continuum states obtained by constraining the system motion to the configuration box.

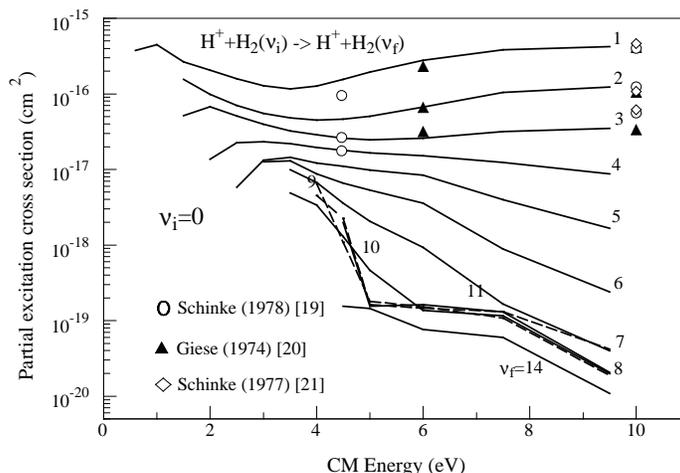


Figure 4. Vibrational excitation cross sections for collisions of protons with H_2 in the ground state.

In addition to a limited number of bound vibrational states (totaling 35 on both H_2 and H_2^+), several hundred continuum states were needed (including the closed channels) to achieve convergence of the cross sections in the energy range considered.

Fig. 4 shows cross sections for excitation by proton impact of the ground state of H_2 to all bound vibrational states. The results are compared with cross sections from the literature. The role of the particle rearrangements in the excitation process is of particular importance at the lowest energies. The previous quantum calculations shown in Fig. 4, do not include the latter channels, underestimating the cross sections at lowest energies. Our data contain coherent sums over the direct and rearrangement channels.

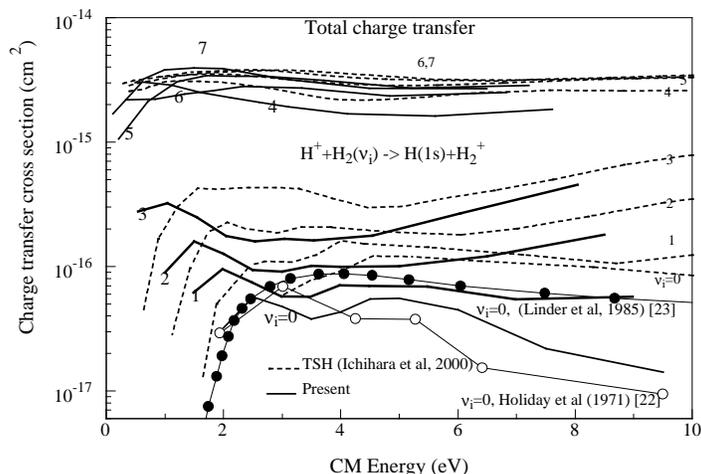


Figure 5. Charge transfer from vibrationally excited states of H_2 .

The charge transfer cross sections (summed over final vibrational states) from initially excited vibrational states of H_2 in collisions with protons are shown in Fig 5. Comparison with experimental data of Holiday [22] and Linder [23] shows good agreement at lowest energies, but lies between these two data sets at the higher energies. Among all the cross sections shown, charge transfer from the first three excited states constitutes a separate group. The reason is that CT from the higher excited states is dominantly exoergic and often quasi-resonant with the vibrational states of H_2^+ . The corresponding cross sections therefore are large even at the lowest energies. Comparison of our data with the TSH ones of Ichihara [17] shows good agreement for the higher initial vibrational states, as could be expected from the correspondence principle. It is interesting to note that our quantum and the TSH calculations show similarly large contributions of the particle re-arrangement channels at low energies.

As a contrast to Fig. 5, Fig. 6 shows charge transfer cross sections from excited states of H_2^+ in collision with H. Since this process is exoergic from all vibrational

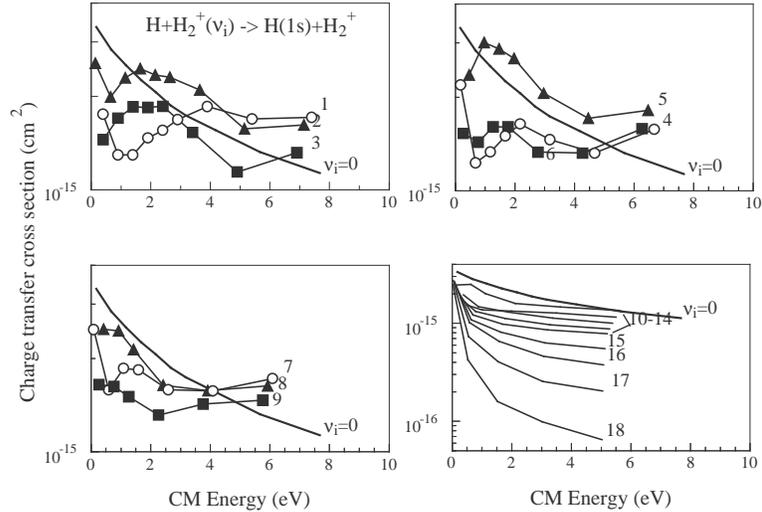


Figure 6. Charge transfer from vibrationally excited states of H_2^+ .

states (including the ground one), the characteristic increase of the cross section toward lower energies is expected, and is not strongly dependent on the initial state. The cross sections from high vibrational states are suppressed due to depletion of these states to the vibrational continuum (dissociation).

The CT cross sections from the 13th excited state of H_2^+ to vibrational states of H_2 in collision $H+H_2^+$ are shown in Fig. 7, for various collision energies. As expected, the distribution peaks around quasi-resonance with the 11th excited state of H_2 . Figure 8 presents the dissociation cross sections, for all initial vibrational states of H_2 . Comparison with the TSH calculations shows a good agreement for higher vibrational states at the higher end of the collision energy range. We note that this process includes both direct dissociation into the dissociative continuum of H_2 , as well as charge transfer dissociation into the continuum of H_2^+ . These two channels are of the same order of magnitude. Concerning the energy spectra of the dissociating fragments,

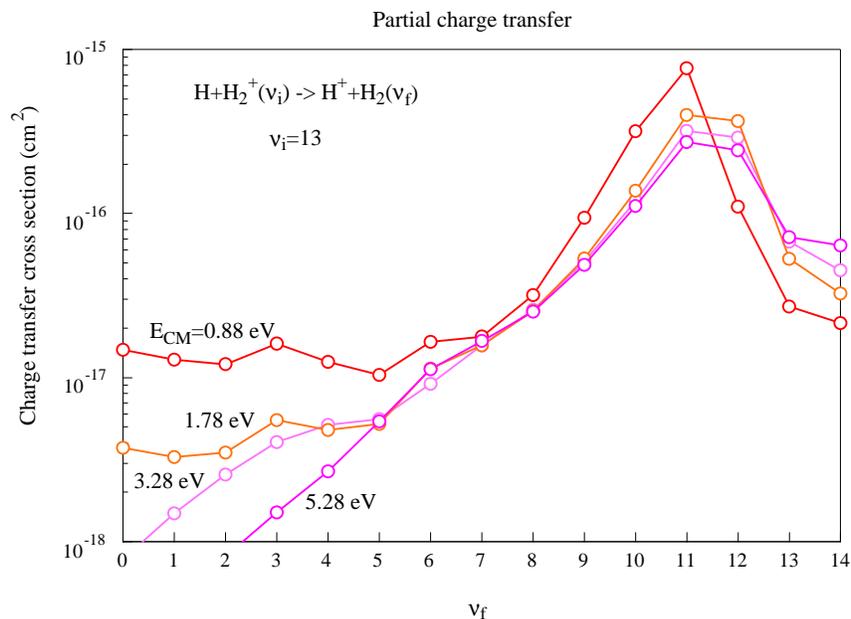


Figure 7. State resolved cross sections for charge transfer to excited states of H_2 .

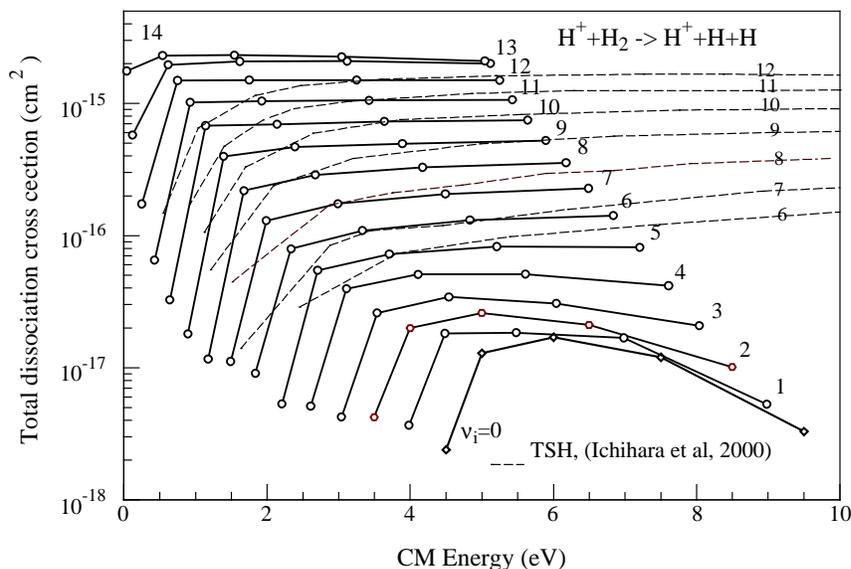


Figure 8. Cross section for dissociation from various excited states of H_2 .

these have the characteristic cusp at the continuum edge, which is more pronounced for the lower collision energies.

The database produced can be found at www-cfadc.phy.ornl.gov and contains all the processes mentioned above for the $H+H_2^+$ and H^++H_2 collision systems in the form of partial and total, initial- and final-vibrational-state-resolved cross sections. This represents currently the most comprehensive set of quantum-mechanically obtained inelastic data for collisions that involve hydrogen atoms, ions and molecules. Still, in order to study rovibrationally resolved processes at low collision energies with various

isotopic combinations of H and, with resolved particle rearrangement channels, the rotational dynamics have to be fully accounted for. This will be the subject of our future work.

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