Electron-impact dissociation of molecular hydrogen: benchmark cross sections

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Electron-impact dissociation of molecules is an important process which plays a major role in governing the dynamics of astrophysical, industrial, and fusion plasmas. Because of its universal abundance, the H_2 molecule is a significant species in many plasma environments, particularly in the divertor region of tokamak reactors, where more than 90% of the neutral hydrogen is molecular [1].

At low energies (below approximately 14 eV), the primary pathway to dissociation of ground-state H_2 is through excitation of the $b\ ^3\Sigma_u^+$ state producing neutral H($1s$) atoms. There has been a long-standing disagreement between theory and experiment for this transition. The current recommended data [2] are derived from measurements more than 20 years old, and various calculations have predicted cross sections both significantly higher and lower than experiment [3]. A theoretical approach to electron impact excitation of the $b\ ^3\Sigma_u^+$ state of H_2 exhibits difficulties characteristic to electron-molecule collisions in general. At energies close to the excitation threshold the effects of nuclear motion have to be taken into account. As incident electron energy increases, inter-channel coupling plays a dominant role, both making theoretical treatments extremely difficult. In addition, the lack of spherical symmetry makes computational treatments particularly expensive.

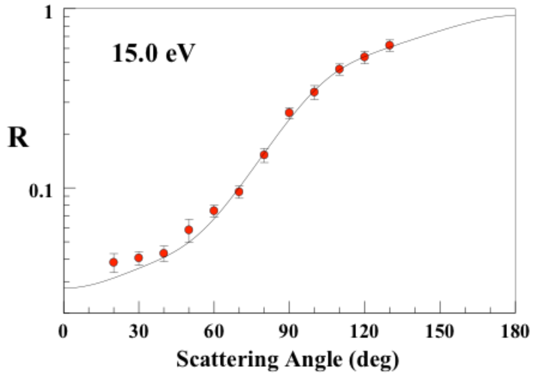

Figure 1: *Ratio R of inelastic to elastic DCS for 15 eV electrons scattering on H_2.*

Recently, the Convergent Close-Coupling (CCC) method has been utilized to provide differential and integrated cross sections for the $b\ ^3\Sigma_u^+$ state of H_2. The CCC method [4] for molecules utilizes large close-coupling expansions to describe e-H_2 collisions. The set of H_2 target states used in such expansions is obtained via diagonalization of the H_2 Hamiltonian in a Sturmian (Laguerre) basis that allows it to model all important reaction channels including ionization. These calculations were performed in the adiabatic nuclei approximation below 14 eV [5] and the fixed-nuclei approximation at higher energies [3]. The CCC results are in good agreement with the recommended data [2] below 12 eV, but predict a cross section up to a factor of two smaller at higher energies.

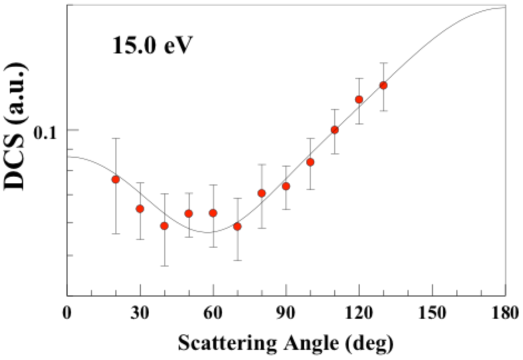

Figure 2: *DCS in atomic units for excitation of b $^3\Sigma_u^+$ state at 15 eV. Comparison is between the present measurements of Khakoo* et al. *[6] and the present CCC results.*

In a joint experimental and theoretical investigation, we have now resolved the discrepancy between experiment and theory for this fundamental process. The California State University (Fullerton) group [6] have recently utilized a newly designed transmission-free electron time-of-flight (TOF) spectrometer to measure differential cross sections (DCS) for the $X\ ^1\Sigma_g^+ \rightarrow b\ ^3\Sigma_u^+$ transition, as ratios of inelastic to elastic scattering. In Fig. 1 we present an example of such ratio measurements and compare the results with the CCC calculations at 15 eV. In Fig. 2 the absolute DCS are presented at the same incident electron energy, and the integrated cross sections (ICS) are presented in Fig. 3 over a range in incident energies. We find outstanding agreement between the CCC calculations and the updated measurements, signifying major progress in electron-molecule scattering and demonstrating the accuracy of the molecular CCC method.

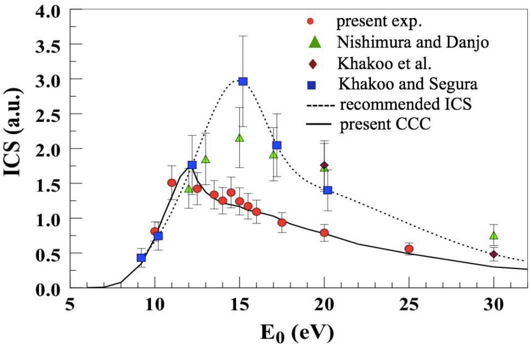

Figure 3: *ICS for excitation of the b $^3\Sigma_u^+$ state of H_2. Comparison is between the previous experiments of Nishimura and Danjo [7], Khakoo* et al. *[8], and Khakoo and Segura [9], the present measurements of Khakoo* et al. *[6], and the present CCC results.*

Utilizing the same molecular CCC calculations [3, 5], we additionally present cross sections for electron-impact dissociation from the H_2 ground state into all neutral fragments $\mathrm{H}(n\ell)+\mathrm{H}(n'\ell')$, for energies from 6 to 300 eV. The CCC calculations [11, 12] account for dissociative excitation, excitation radiative decay dissociation, and predissociation through all bound electronic triplet states, and singlet states up to the $D'\ ^1\Pi_u$ state. An estimate is given for the contribution from the remaining singlet states. Previous calculations of dissociation into all neutral fragments has previously been unfeasible due to the need to take account of a large number of excited electronic states.

Below approximately 20 eV, excitation of the repulsive $b\ ^3\Sigma_u^+$ state makes the dominant contribution to the dissociation cross section. Above this energy, the remaining triplet-state excitations make an approximately equal contribution to the $b\ ^3\Sigma_u^+$ state towards dissociation (see Fig. 4). Since radiative decays must preserve the target spin, the triplet states cannot decay back to the ground state, and hence all excitations in the triplet system lead to dissociation. The cross section for dissociation through the triplet states is therefore obtained by summing the respective excitation cross sections. We utilize the adiabatic-nuclei calculations discussed above for the $b\ ^3\Sigma_u^+$ state, but find the fixed-nuclei results to be sufficient for all other transitions as the $b\ ^3\Sigma_u^+$ state is dominant at the threshold energies of the higher states.

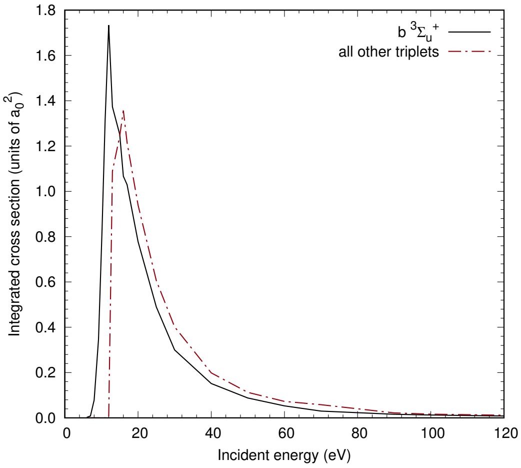

Figure 4: *Cross sections for dissociation through excitation of the* $b\ ^3\Sigma_u^+$ *state, and all other triplet states.*

Above about 35 eV, the singlet-state excitations overtake the triplet states as the dominant dissociation pathway. The $B\ ^1\Sigma_u^+$, $C\ ^1\Pi_u$, $B'\ ^1\Sigma_u^+$, $D\ ^1\Pi_u$, $B''\ ^1\Sigma_u^+$, and $D'\ ^1\Pi_u$ *ungerade* singlets make a significantly larger contribution than the remaining singlet states. We obtain a total cross section for dissociation into neutral fragments by summing the cross sections for excitation of all bound electronic triplet states, and the weighted cross sections for the singlet-state excitations. The dissociation cross section is demonstrably convergent with respect to the number of coupled channels, and we have provided an uncertainty estimate which accounts for the numerical convergence and target structure, as well as the present treatment of higher singlet states. The recommended data for this process produced by Yoon *et al.* [2] were inferred from the total dissociation yield (including charged fragments) measured by Corrigan [10] in 1965, by subtracting the recommended ionization cross section. In the subsequent 50 years, there have been no attempts to repeat these measurements, and to

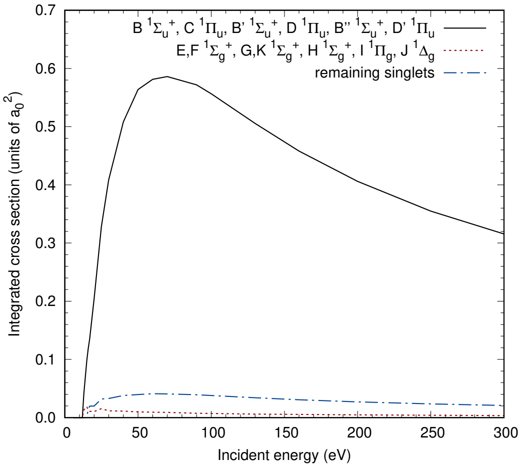

Figure 5: *Cross sections for dissociation through excitation of a number of gerade and ungerade singlet states, with an estimate for dissociation through the remaining singlet state we do not treat explicitly.*

date there have been no direct measurements of dissociation producing only neutral fragments. Previous theoretical treatments have been limited to low energies where only a small number of dissociation channels are open, or they have only considered dissociation producing hydrogen atoms in a limited range of quantum states. Our present CCC results are in good agreement with the recommended data in the low (6–12 eV) and high (60–70 eV) energy regions, but somewhat lower at the intermediate energies. The uncertainties in our results, while likely overestimated, do not account for the discrepancy with the recommended data. This suggests that new measurements over the entire range of impact energies are highly desirable.

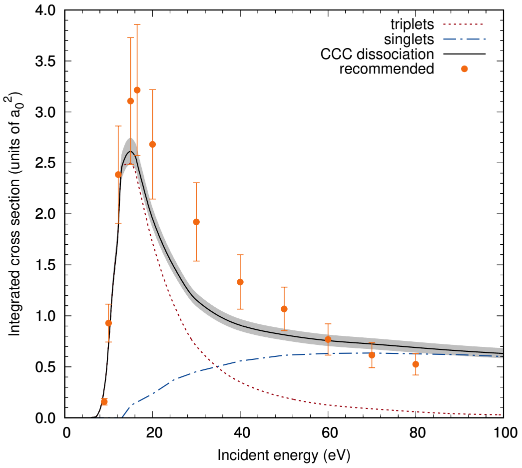

Figure 6: *Electron-impact dissociation cross section calculated in the CCC method compared with the recommended data [2]. The shaded region indicates the uncertainty in the present results.*

The comparison of the present results with the currently recommended data, and the lack of any other previous calculations, mean that new measurements of the H_2 dissociation cross section are highly desirable. We expect that the present calculations will be of interest for modeling astrophysical and fusion plasmas, where the hydrogen molecule is abundant.

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