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Calculation of Low-Energy Electron Dissociative Attachment of Molecular Hydrogen for Plasma Applications

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Abstract

Calculation of rate constants for dissociative electron attachment to molecular hydrogen is reported. The calculation is based on an improved nonlocal resonance model of Čížek, Horáček and Domcke which takes fully into account the nonlocality of the resonance dynamics and uses potentials with correct asymptotic forms. The rate constants are calculated for all quantum numbers v and J of the target molecules and for electron temperature in the range 0 – 30 000 K.

Keywords:

dissociative attachment, molecular hydrogen, rate constant

1. Introduction

The system H_2^- is the most fundamental molecular anion. It is unstable, with a very short lifetime ($\tau \sim 10^{-15}$ s). The dynamics of this complex which is important in a number of collisional processes is still not well understood. As examples of the most studied processes we mention dissociative attachment (DA)

$$e^- + H_2 \rightarrow H + H^-, \qquad (1)$$

vibrational excitation (VE)

$$e^- + H_2(v_i) \rightarrow e^- + H_2(v_f), \qquad (2)$$

associative detachment (AD)

$$H + H^- \rightarrow H_2 + e^- \qquad (3)$$

and, for scattering energies higher than the electron affinity of H, also collisional detachment (CD)

$$H + H^- \rightarrow H + H + e^-. \qquad (4)$$

Understanding of these processes is important for a number of practical applications. AD and DA determine the thermal equilibrium densities of H^- ions and H_2 molecules in many astrophysical plasmas. Neutral molecules produced in AD are vibrationally excited and the emission spectra of such molecules are quite different from those of molecules excited by ultraviolet pumping or shock excitation [1]. DA of electrons to molecular hydrogen is thought to be the primary source of the H^- ions produced in hydrogen plasmas. These ions may serve for the generation of neutral particle beams, the injection of ions into controlled thermonuclear devices, or for electromagnetic propulsion of space vehicles.

In the early models of the H_2^- dynamics the interaction between the nuclei was described in the local-complex-potential approximation [2-4] with empirically adjustable parameters and later with *ab initio* potentials [5, 6]. The importance of nonlocal effects was pointed out in [6]. Recently, the dynamics of nonlocal models has been studied [7-13], for DA and VE in H_2.

The present work is based on an extension of the nonlocal resonance theory to treat the DA to rotationally excited molecules (for review of the nonlocal resonance model see [14]). The existing nonlocal resonance model for the H_2^- dynamics [7, 9] is modified [15] to account for the long-range behaviour of the H_2^- potential-energy function. DA, VE and AD processes can thus be treated within a single model with no adjustable parameters.

2. Dissociative attachment cross sections

The main emphasis of this work is on the calculation of rate constants for DA process. It is however instructive to discuss briefly the underlying DA cross sections.

In Fig. 1, the calculated DA cross sections are plotted assuming nonrotating ($J = 0$) molecular target for several values of the vibrational quantum number v. A

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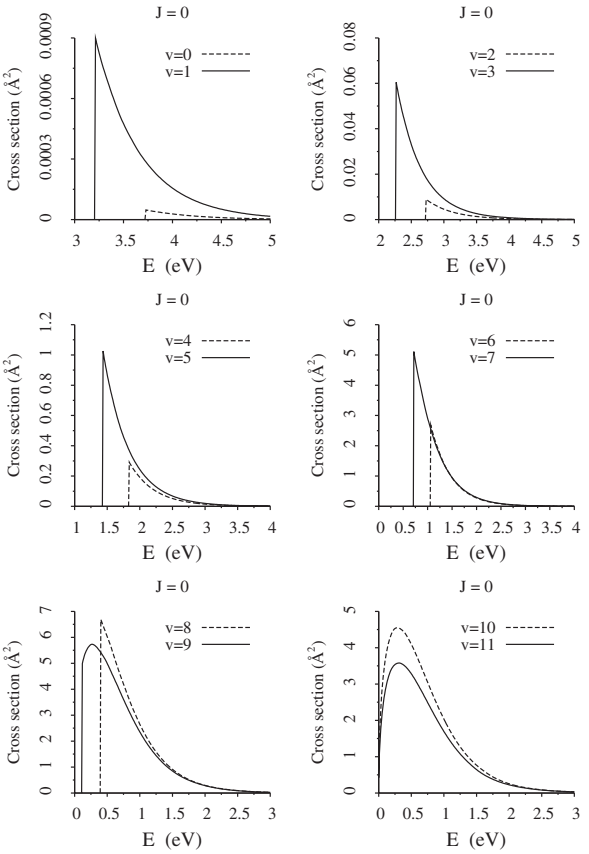

Fig. 1 DA cross section for initial vibrational target states $v = 0, 1, \ldots, 11$ for nonrotating hydrogen molecule.

typical feature of DA process in molecular hydrogen is the rapid increase of the cross sections with increasing vibrational energy of the target which determines the strong temperature dependence of the cross section. This feature is clearly demonstrated in Fig. 1. The magnitude of the cross section increases at increasing vibrational excitation of the target v up to $v = 8$. This feature is used for example as a diagnostic tool in [16].

It is generally accepted that the effect of rotational excitation on the DA cross section is comparatively small and that the effect of vibrational excitation is responsible for the observed increase with the temperature. This is true only, however, at low values of the rotational quantum number J. With increasing J, the DA cross section changes its form; the onset becomes smooth and the cross sections attain their maxima at energies above the threshold. With increasing J, the rotational heating becomes more efficient than the vibrational heating. The largest DA cross section predicted by the present model arises for $v = 1$, $J = 29$ with a peak value of 28.3 Å^2 at $E = 134$ meV. The largest cross section for an endothermic DA process is obtained for $v = 2$, $J = 23$. This reaction opens at 113 meV and the cross section reaches the value of 20.1 Å^2 at 249 meV. For more details see [17].

The peak value of the DA cross section at low energies is a very important quantity and in fact some theories were adjusted to it [4]. The magnitude of the cross section represents still an open problem. Two values exist in the literature: The value of 2.8×10^{-5} Å^2 obtained by Schulz and Asundi [18] and 1.6×10^{-5} Å^2 by Rapp *et al.* [19]. The measurement of Schulz and Asundi was done at a temperature of about 300 K with an energy resolution of about 450 meV. If our cross section is evaluated at this temperature and convoluted with the assumed experimental energy resolution we obtain a value of 2.85×10^{-5} Å^2, in full agreement with the measurement of Schulz and Asundi.

3. Dissociative attachment rate constants

The dissociative attachment constants are needed for modelling various plasma processes. Generally, the rate constant for the reaction in gas is defined as

$$k = \int_0^\infty v f(v) \sigma(v)\, dv \qquad (5)$$

where $f(v)$ is a normalized Maxwellian distribution of the relative velocities v and $\sigma(v)$ is the cross section of the reaction.

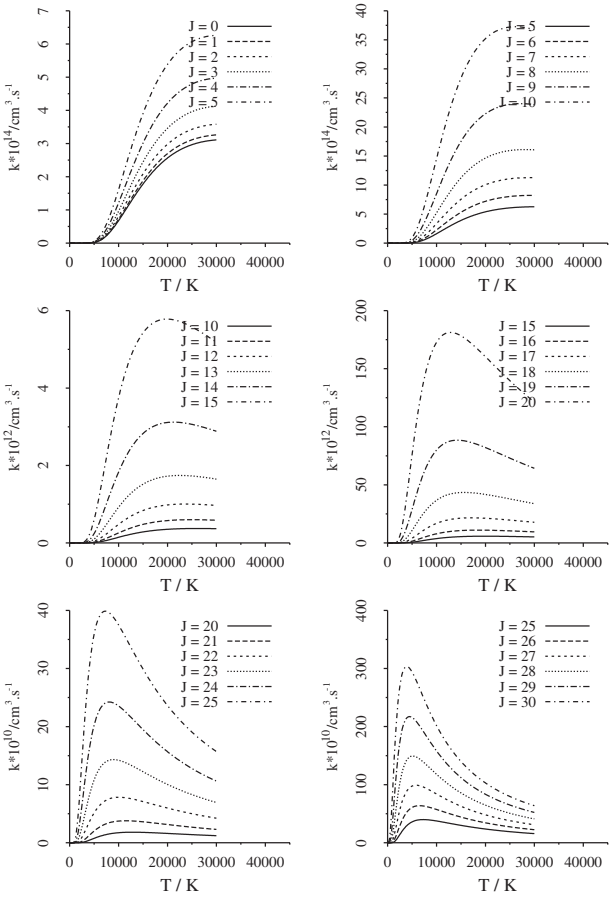

Fig. 2 DA rates for ground vibrational state $v = 0$ and various rotational states J.

The calculated rate constants are plotted in Fig. 2 as functions of the electron temperature T for hydrogen molecules in their ground vibrational state, $v = 0$, and various rotational states $J = 0, 1, \ldots, 30$. It is seen that the rate constants are strongly temperature dependent. Using these data the DA rate constant assuming the gas temperature T_g can be easily calculated. For details see NIFS-DATA-73.

4. Discussion

The nonlocal resonance model is the most successful method to treat resonance inelastic processes in low-energy electron-molecule collisions. It reproduces all features observed in measured cross sections, namely the threshold peaks in the vibrational excitation of hydrogen halides, Wigner cusps, oscillatory structures in elastic and inelastic cross sections converging to the dissociative attachment threshold, isotope effect, temperature dependence of the cross sections, etc. Moreover, the application of the nonlocal resonance model to highly rotationally excited hydrogen molecules led to the discovery of very long-lived states of H_2^-. The resonance states of molecular hydrogen anions are known to be of very short life-time of the order of 10^{-15}s. The nonlocal resonance model however predicts the life-time of hydrogen anions of the order of microseconds for highly rotationally excited molecules ($J \approx 23$). The existence of the predicted long-lived states was confirmed experimentally in the accelerator mass spectrometry [20]. Even though the electron-hydrogen scattering process has been studied in various ways no approach has ever predicted the existence of these states. The present model has however limitations. Since it takes into account only the $^2\Sigma_u^+$ resonance it is good only at low energies below 5 – 6 eV. As all resonance models it ignores the direct scattering contribution which is however assumed to be very small in VE and DA processes.

In plasma physics the local complex potential approach is used for the calculation of DA and VE cross sections. The models are usually obtained by fitting several parameters to experimental DA data. This approach however fails seriously for the $^2\Sigma_u^+$ shape resonance in e + H_2. Neither the magnitude nor the shape of the vibrational excitation cross section is correctly reproduced by the local complex potential approximation [9]. The present model which has no adjustable parameters yields vibrational excitation cross sections in very good agreement with experimental data. This is shown in Fig. 3 where the present data for the $0 \rightarrow 1$ VE are compared with the experimental data [21]. Nonlocal effects are essential for the correct description of

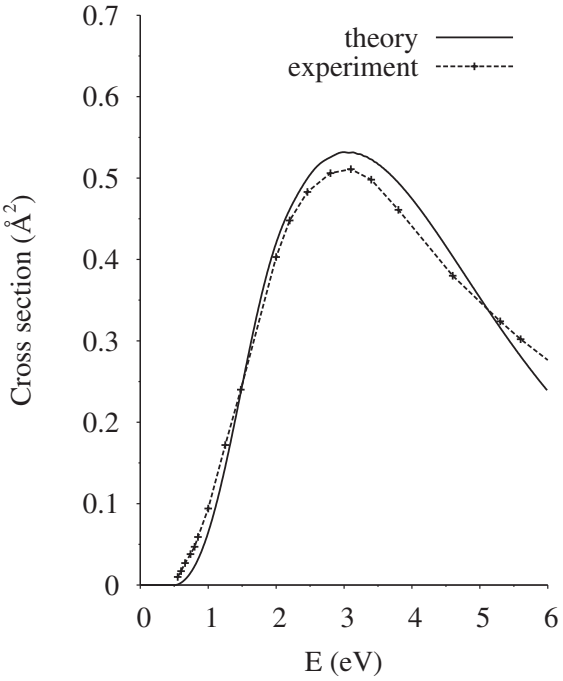

Fig. 3 Calculated VE $0 \rightarrow 1$ cross section - solid line - compared with the measurement of Allan [21] - crosses.

inelastic processes in H_2 for low vibrational and rotational states of the target.

It is the purpose of this short contribution to show that the improved nonlocal resonance model [15] can produce also data for molecular hydrogen useful for the modelling various plasma processes. Besides the data on DA rate constants discussed here the model yields also vibrational excitation cross sections and cross section for associative detachment. A detailed evaluation of the computed data and their comparison with experimental data and other calculations can be found in [17] and will be omitted in this short contribution. The DA rate constants were calculated at a range of electron temperatures $T = 0 - 30000$ K for hydrogen molecules for all rotational and vibrational states (v, J) of the hydrogen molecules. Sample results are shown in Fig. 2. The complete data are obtainable from NIFS data base (*e.g.* NIFS-DATA-73).

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