Collisional-radiative model of Nanosecond Repetitively Pulsed Discharges and Dielectric Barrier Discharges in hydrogen plasmas

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Abstract

The role of vibrational kinetics of H_2 in high pressure pulsed discharges is discussed. To this end, a self-consistent model coupling chemical kinetics, internal distributions and free electron kinetics has been used to perform 0D time dependent calculations. This approach allowed to investigate the complex interaction between non equilibrium distribution of internal levels and of free electrons, these last strongly affected by the presence of superelastic collisions with both vibrational and electronically excited states.

Introduction

Atmospheric pressure non-thermal discharges are promising technologies with many practical applications such as a) aerodynamic flow control through electro hydrodynamic interaction b) combustion enhancement, flame stabilization c) plasma destruction/activation of greenhouse gases d) surface treatment/modification including the creation of nanoscale structures e) diamond nanoparticle synthesis.

Hydrogen is widely used as process gas in plasma treatments, and a lot of work is currently being devoted to fundamental investigations of low-pressure hydrogen plasmas due to their technological importance in photovoltaics, deposition of diamond films, aerospace, nuclear fusion and negative ion generation.

The role of vibrational excitation in high pressure discharges is often underestimated, because the characteristic time of VT processes is much longer than that of the discharge. Moreover, electron induced processes included in the calculations involve only excitation from the ground vibrational state. This work addresses the study of nanosecond-pulsed discharges in pure hydrogen at atmospheric pressure focusing on the interaction of the vibrational distribution (VDF) with electron kinetics and chemical composition.

Model description

The numerical model used in this work is based upon our previous work [1, 2, 3], improved taking into account an extended kinetic scheme for the H_2 plasma and coupling the 0D kinetics to the equations of an external circuit, to model the ns-RPD and DBD conditions.

In brief, the model includes

i) a set of master equations describing the time evolution of the gas composition and internal distributions of atomic and molecular species

ii) the Boltzmann equation for the electron energy distribution function, EEDF, taking into account an external electric field

iii) the equation for an RC circuit chosen to determine the *E-I* characteristic of the discharge.

The set of master equations refers to a pure hydrogen gas considering H_2, with 15 vibrational levels and 8 electronic states ($B^1\Sigma_u^+$, $C^1\Pi_u$, $D^1\Pi_u$, $B'^1\Sigma_u^+$, $B''^1\Sigma_u^+$, $D'^1\Pi_u$, $a^3\Sigma_g^+$, $c^3\Pi_u$), H_2^+, H, with the ground and 9 excited states, H^+, e^-, with a total 34 pseudo-species.

The full description of processes considered and the set of cross sections and rate coefficients used is given in refs. [4, 3]. In order to better describe the present high pressure discharge conditions, the H_3^+ and H^- ionic species have been included in the kinetic scheme.

The H^- ion is mainly produced by dissociative attachment to vibrationally excited H_2

$$e + H_2(\upsilon) \rightarrow H(n=1) + H^-$$

and then consumed by several neutralization reactions with positive ions [5, 6]. The H_3^+ molecular ion is mainly produced in the reaction [7]

$$H_2 + H_2^+ \rightarrow H_3^+ + H$$

and then destroyed by recombination and ion conversion processes. The characteristics of a discharge depend not only on the applied field but also on the polarization circuit, influencing the ionization degree, chemical composition and excited states population. A general approach to these kind of discharges can be to to consider a series RC circuit (figure 1), solving the circuit equation to get the current though the plasma, which is considered as a purely resistive element. The effect of surface charge accumulation in the DBD case has been accounted for introducing a capacitance C in series with the circuit.

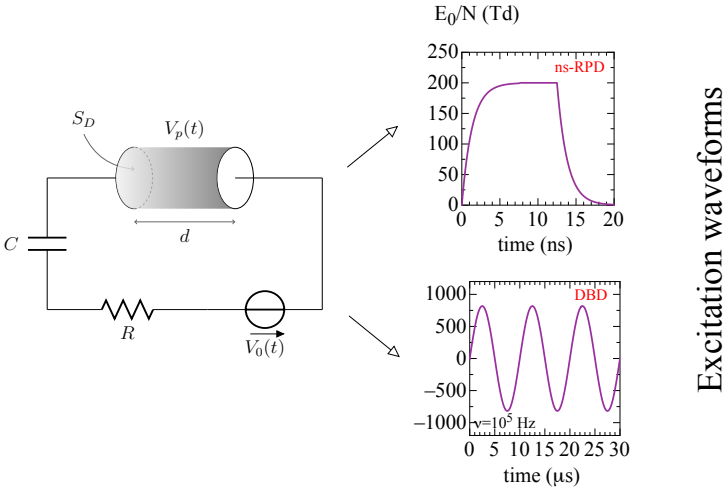

Figure 1: Circuit scheme.

Results

In this section we present the results for the ns-RPD in pure H_2 molecular gas. The discharge is generated by an elecric field pulse $E_0(t)$ with finite raise and fall times, see figure 1.

Figure 2 reports the time evolution of the molar fractions of chemical species along a time interval corresponding to 6 cycles discharge–postdischarge. It can be observed that in the first voltage the gas composition remains almost constant and its main effect is initiating the H_2 ionization, increasing and heating the ambient electrons. Electron temperature grows very rapidly, so that ionization is the preferred channel. Chemical processes and electronic excitation are more favoured in these conditions. Starting from the third pulse, the plasma shows periodic behaviors. The main ionic species in between pulses are H_3^+ and H^-, while during the pulse time the main species is H^+.

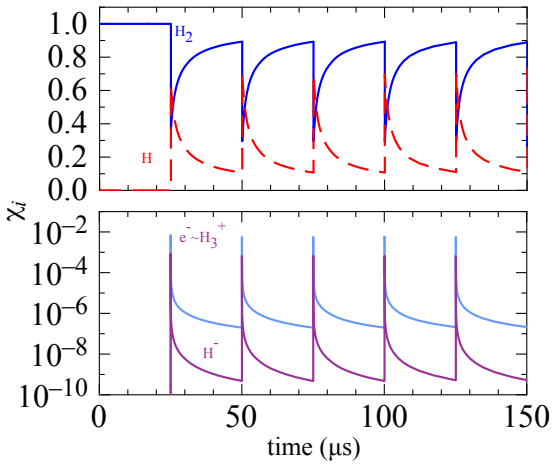

Figure 2: Neutral and ionic concentrations during the first 6 pulses.

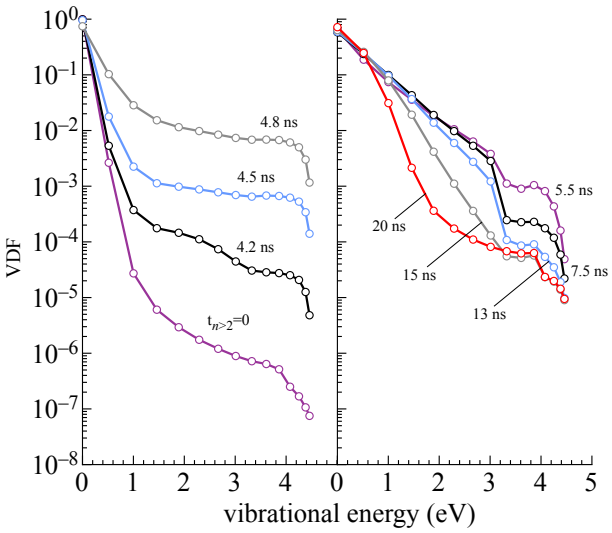

Figure 3: Vibrational distribution during the third pulse.

Evolution of H_2 vibrational distribution during the third pulse is reported in figure 3. At the beginning of the pulse the VDF presents a minimum and a maximum, behavior due to the incompleteness of the eV cross section set. Up to 4.8 ns from the pulse start, the vibrational distribution is heated, with the last levels underpopulated depleted by VT processes. Then, while the low energy distribution is heated by electrons, reaching the equilibrium between VT depletion and electron impact excitation, the high energy distribution decreases, balancing between excitation, VT, dissociation, ionization and recombination. Then a quasi-steady state distribution is obtained until the beginning of the field falling phase, where the distribution is cooled down. To show the relevance of transitions from vibrationally excited states we are comparing the re-

sults including transitions from each vibrational state ($\upsilon = all$) with those obtained considering only transitions from the ground state ($\upsilon = 0$). The tail of vibrational distribution (see Fig. 4) differ of more than three orders of magnitude: the distribution in the case ($\upsilon = all$) are much higher, due to the larger number of processes than in the case ($\upsilon = 0$). As a consequence the case ($\upsilon = all$) results in higher dissociation.

Acknowledgements

This work received funding from the project 'Apulia Space', PON 03PE-00067.6 from Distretto Tecnologico Aerospaziale Puglia S.C.A.R.L. (Italy).

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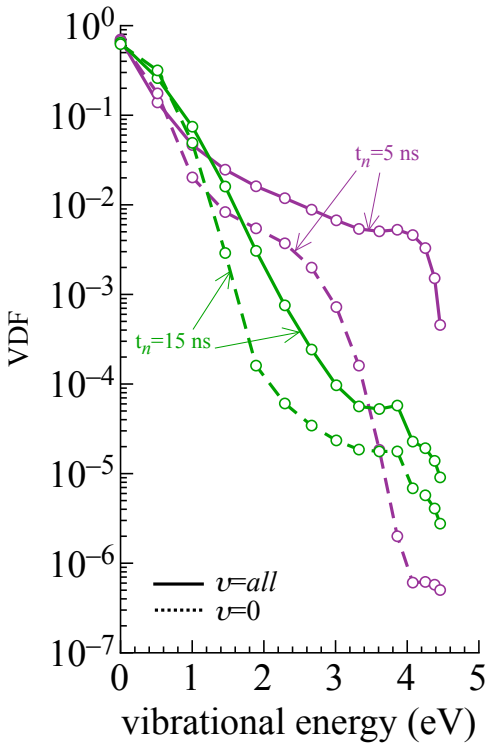

Figure 4: Vibrational distribution during the third pulse comparing the $\upsilon = 0$ to the $\upsilon = all$ cases.