Kinetics of $N_2(B^3\Pi_g)$ and $N_2(C^3\Pi_u)$ states in N_2-Ar discharges sustained by a RF helical coupling device

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A kinetic model is discussed for the two main emitting species (N_2(B), N_2(C)) of a N_2-Ar plasma excited at 27 MHz by a helical cavity with operating pressure of 200 Pa. The present paper consistently illustrates, for miscellaneous Ar fractions and different positions, the great influence of the processes involving the metastable species (N_2(A), Ar^m) and the electrons on the kinetics.

Introduction. Nitrogen containing plasmas are implemented in many applications and industries. The knowledge of the main reactions in such media is essential to achieve their better controllability. In this context, optical emission spectroscopy is carried out to study N_2(B) and N_2(C) states emissions (denoted 1^+ and 2^+, respectively) in a N_2-Ar discharge excited by a RF helical cavity. A global model is built to explain the plasma kinetics through these species.

Experimental section. The N_2-Ar mixture ($0 \leq$ % Ar ≤ 95) is excited at 200 Pa by helical cavity at 27 MHz with a power fixed at 28 W, in a 18 mm inner diameter tube (Fig. 1). The plasma emissions in the range 215 – 950 nm are collected through an optical fiber set into the holes (the second hole, as $z = 0$ cm, is the input power place) analyzed by an Andor Mechelle ME5000 spectrometer ($\lambda/\Delta\lambda = 4000$) coupled with an Andor iStar camera. Fig. 2 shows, as an example, the axial profile of 2^+ as a function of gas mixtures. Similar behavior is observed for 1^+. These profiles can be described by sinusoidal curve according to a nonlinear behavior characteristic of our discharge [1]. The increase of Ar fraction reduces this effect reflecting a probable impact on the kinetics.

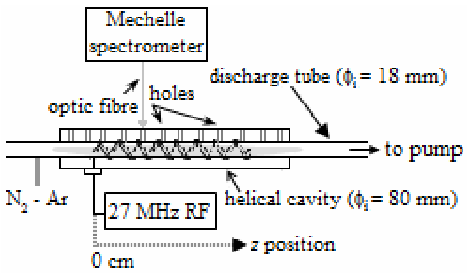

Fig. 1: Experimental set-up.

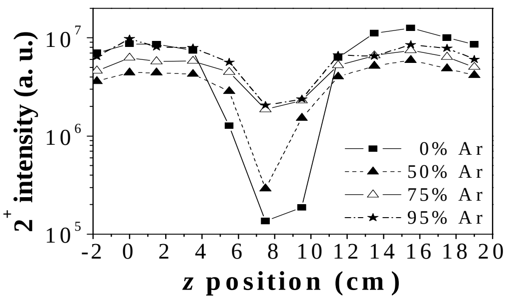

Fig. 2: Axial profiles of 2^+ emissions for different Ar fractions.

Kinetic model. The coupled system of rate balance equations is numerically solved included excitation by electron impact, collisional processes with heavy particles, excitation transfer by the Ar metastable (Ar^m) specie and radiative cascade according to previous works [2]. The input parameters are: gas temperature measured from 1^+ ($T_g = 430$ K) [1], concentration (n_e) and temperature of electrons (T_e), and vibrational temperatures T_v(X) (T_v(A)) for N_2(X) (N_2(A)) states. We assume a Maxwellian Electron Energy Distribution Function while (i) n_e, T_e increase with Ar fraction, (ii) T_e is invariant whereas n_e has a hollow profile along z. T_v(X) and T_v(A) are considered constants along the discharge for all gas mixtures with a value of 5200 and 2000 K, respectively, while T_v(A) shows a weak influence in the model. Moreover, according to our analysis, the effect of nitrogen atoms gives us just a corrective factor, so the density ratio $[N(^4S)]/[N_2]$ is estimated to be constant whatever the Ar fraction with a value of 0.1 %.

Results and discussion. We reproduce with a satisfactory agreement the N_2(B) and N_2(C) populations evolution as a function of Ar fraction whatever the axial position. Fig. 3 presents our results for three z positions. The N_2(B) density increases with Ar concentration up to 90 % and a slight decrease is observed for a 10 % N_2 – 90 % Ar mixture. The $N_2(C)$ density evolution with Ar amount presents two behaviors according to the position: (i) an increase with the Ar fraction at z = 7.5 cm, (ii) a slight decrease up to 50 % Ar, than an increase with rising Ar percentage in the most intense discharge regions (z = 0 and 3.5 cm). The best fit of the experimental data by the kinetic model involves values of T_e ranging from 1.0 to 1.1 eV for all z positions (see Fig. 4 (a)). Furthermore, the n_e evolution, as previously mentioned, is similar to T_e (i.e. with a minimum value located at z = 7.5 cm and reaching a maximal value at z = 0 cm). The evolution of $N_2(A)$ density versus Ar percentage deduced from the model is shown on Fig.4 (b). The density evolution of Ar^m atoms shows a trend similar to that of $N_2(A)$ and suggests that these states should be mainly produced by electron impact. All the densities increase with Ar percentage.

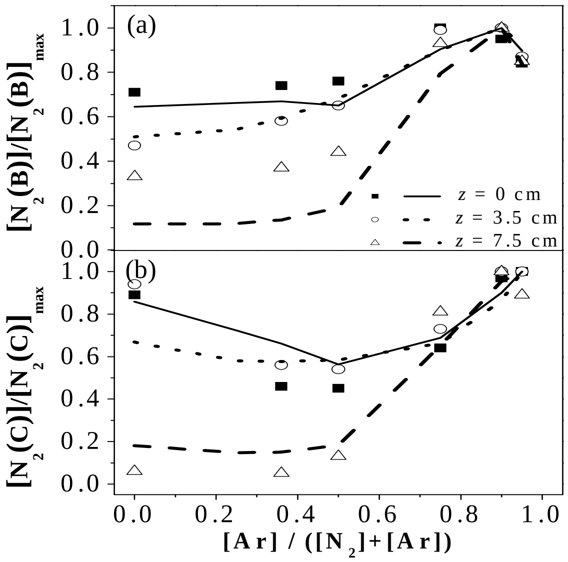

Fig. 3: Comparison of the $N_2(B)$ (a) and $N_2(C)$ (b) densities variations, with respect to their maximum, versus the gas composition for several z values given by experiments (symbols) and kinetic model (lines) .

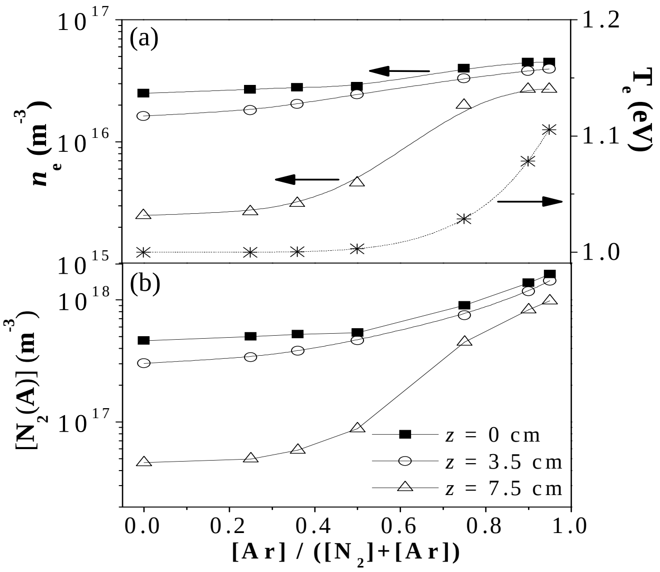

Fig. 4: Variations of (a) n_e and T_e (stars) and (b) $N_2(A)$ state density deduced from the kinetic model versus Ar percentage for several positions.

With regard to our kinetics investigation, we succeed to determine the main mechanisms controlling the $N_2(B)$ and $N_2(C)$ states:

$$e + N_2(X) \rightarrow e + N_2(B, C) \quad (R_1)$$

$$e + N_2(A) \rightarrow e + N_2(B, C) \quad (R_2)$$

$$N_2(A) + N_2(X, v = 5 - 14) \rightarrow N_2(B) + N_2(X), \quad (R_3)$$

$$N_2(A, v = 0, 1) + N_2(A, v = 0, 1) \rightarrow N_2(C) + N_2(X), \quad (R_4)$$

$$N_2(X) + Ar^m \rightarrow N_2(C) + Ar. \quad (R_5)$$

In N_2 discharge, $N_2(B)$ is mainly produced by the (R_3) reaction. With Ar increase, its excitation by electrons from $N_2(A)$ become significant ((R_2) process). The (R_1) direct electron impact contributes to around 20 % of $N_2(B)$ production whatever the gas mixture. On the contrary, (R_1)-type process is predominant in the production of $N_2(C)$ in N_2 plasma. At 95 % Ar, the (R_4) pooling mechanism contributes to more than a half to the overall $N_2(C)$ concentration. At higher Ar fractions, electron impact with $N_2(A)$ and energy transfer from Ar^m (processes (R_2) and (R_5)) form $N_2(C)$ within around 10 % for each of the reaction. The model reveals a great importance of metastable species along with electron impact reactions, which is consistent with theoretical models developed until now [2]. Also Ar addition affects both n_e and T_e and promotes the processes involving $N_2(A)$ species. Furthermore, in a second contribution to this conference [3], the determination and modeling of the $N_2(C)$ vibrational distribution function, using the present results, is discussed and shows also the crucial effect of $N_2(A)$ metastable.

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References

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