Vibrational Distribution Function of $N_2(C^3\Pi_u, v')$ state in N_2-Ar discharge created by a RF helical coupling device

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The Vibrational Distribution Function of the N_2(C, v'=0-4) state is analyzed by using optical emission spectroscopy in N_2-Ar (0-95% Ar) discharge induced by a helical cavity excited at 27 MHz and 200 Pa. The state-to-state modelling shows that the kinetics involving N_2(A) metastable specie becomes dominant with increasing the Ar amount in the gas mixture.

Introduction. This study is a continuation of our work started within an investigation in terms of kinetic processes of a N_2-Ar discharge created by a RF helical coupling device [1]. The Vibrational Distribution Function (VDF) of the N_2(C) have been determined by means of optical emission spectroscopy. Additionally, the modelling of vibrational distribution of this state is developed on the basis of predictions of the global model [1].

Experimental section. The experimental apparatus is the same as that mentioned in an other contribution of this conference [1]. The N_2-Ar mixture ($0 \leq$ % Ar ≤ 95) is excited at 200 Pa by helical cavity at 27 MHz. To study the emission, twelve holes are made along the cavity. The position of the second hole (z = 0 cm) corresponds to the place where the RF power (28 W) is supplied to the plasma. The VDF of N_2(C) state is deduced from the intensity of the second positive emission system recorded in the range 290 – 420 nm by an Andor Mechelle ME5000 spectrometer ($\lambda/\Delta\lambda$ = 4000) coupled with an Andor iStar intensified camera. The population of the N_2(C) molecule in the v' level, [C,v'], is related to the intensity, $I_{v'v''}$ by the following relation:

$$I_{v'v''} = c(\lambda)\frac{[C,v']A_{v'v''}}{\lambda}$$

where $c(\lambda)$ is the spectrometer response factor for the wavelength λ and $A_{v'v''}$ the Einstein coefficient for the spontaneous emission given by [2].

Tab. 1. Densities and temperatures in the plasma deduced from [1] for the position z = 0 cm

% N_2 – % Ar	Densities of *i* specie, [*i*] (10^{16} m^{-3})			Temperature* (K)			
	[e]	[N_2(A)]	[Ar^m]	T_{gas}	T_e	T_v(X)	T_v(A)
100 % – 0 %	2.5	46	0	430	11600	5200	2000
5 % – 95 %	4.5	160	0.13	430	12750	5200	2000

***T_e** is the electron temperature, T_v(X) (T_v(A)) is the vibrationnal temperature of N_2(X) (N_2(A)) specie.

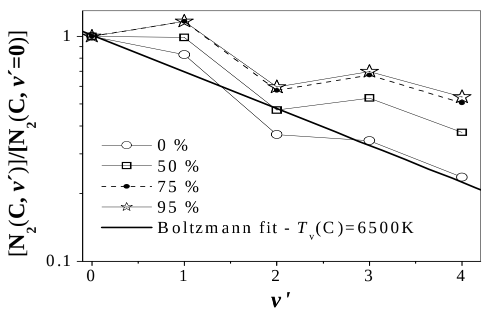

Fig. 1: Experimental VDF of N_2(C,v'=0-4) versus % Ar

State-to-state modelling of the $N_2(C^3\Pi_u, v'$=0-4) specie. The VDF model is built consistently with our global model [1] which demonstrates the great importance of the electron and N_2(A) state in the kinetic of N_2(C). Tab. 1 summarizes the main discharge quantities derived from this model. Based on the literature [3], the considered reactions are listed in Tab. 2. In this table, the sets of coefficients tested for the pooling reaction (R_3) are mentioned.

Results and discussion. Fig.1 presents the N_2(C) VDFs as a function of Ar amount. In N_2 plasma, the VDF follows roughly a Boltzmann-like distribution and the vibrational temperature of the N_2(C) state (T_v(C)) deduced is 6500 ± 1000 K. While increasing the Ar percentage, some departures from Boltzmann plot appear for the levels *v*' = 1, 3 and 4. To our knowledge this is the first time that such deviations are observed. Fig. 2 shows the comparison between the experimental data and those given by the model for the two extreme gas mixtures. In N_2 discharge, the differences between the fits are small. It demonstrates that the N_2(C) VDFs is not sensitive to the effect of the pooling reaction. As shown in Fig. 3, the N_2(C, *v*') state is mainly produced by direct electron impact from the N_2 ground state (process (R_1)). For the 5 % N_2 – 95 % Ar discharge, the overpopulations for *v*' = 1, 3 and 4, with respect to a Boltzmann distribution observed in the N_2(C) VDFs, involves the effect of the (R_3) reaction in the kinetics of the considered specie. The (R_3) coefficients calculated from a Franck-Condon (FC) model (case (b)) cannot reproduce the shape of experimental VDF. This fact does not agree with the observations made by [3] and confirms that the excitation coefficients of the N_2(A) energy pooling cannot be predicted with the FC schemes [4]. We reproduce with a satisfactory agreement the N_2(C) VDF in our N_2-Ar discharge by using the coefficients given by Piper for the (R_3) reaction (model (a) on Fig. 2). But the fitting is questionable for the level *v*' = 2 which appears underpopulated with respect to the others levels. This can be corrected by arbitrary changing two Piper's coefficients within the limits of 30 % error mentioned by this author for the total coefficient (see Tab. 2.).

Tab. 2. Rate coefficients for the reactions considered in the kinetic model

Reaction	Rate coefficients	Ref.
(R_1) e+N_2(X,*v*)→e+N_2(C,*v*') (R_2) e+N_2(A,*w*)→e+N_2(C,*v*')	$k^{v'}_{X,v\ \mathrm{or}\ A,w} \propto \sigma^{C}_{X\ \mathrm{or}\ A} \times q^{C,v'}_{X,v\ \mathrm{or}\ A,w}$ (i.e. cross section×Franck-Condon factors (FCF))	[3]
(R_3) N_2(A,*w*)+N_2(A,*w*') ↳ N_2(C,*v*')+N_2(X)	$k^{v'}_{w,w'}$ from (a) [4], (b) a FCF model, and (c) [4] with modifications as $k^{1}_{0,0}$ =5.3×10^{-17} m^3 s^{-1} and $k^{2}_{0,0}$ =2.9×10^{-17} m^3 s^{-1} (presently 4.1×10^{-17} m^3 s^{-1} for both coefficients)	
(R_4) N_2(X, 0)+Ar^m→N_2(C,*v*')+Ar	$k^{v'}_{\mathrm{Ar^m}}$	[5]
(R_5) N_2(C,*v*')→N_2(B,*v*'')+$h\upsilon$(2^+)	$k^{v'}_{\mathrm{rad}}$	[2]

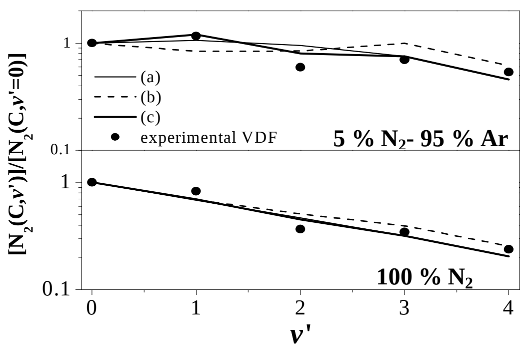

Fig. 2: VDF of N_2(C, *v*'=0-4) state fitting by kinetic models (see Tab.2)

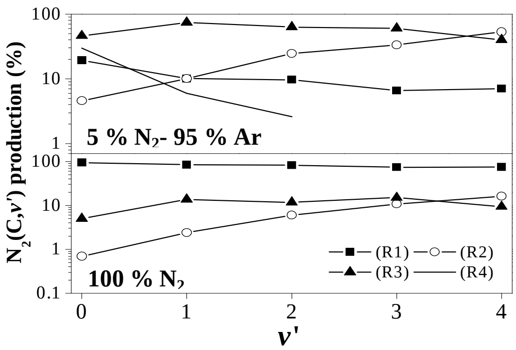

Fig. 3: Contribution of source channel relative to N_2(C, *v*'=0-4) state (case (c))

We conclude that only the two first vibrational levels of N_2(A) state are involved in the N_2(C) production, in agreement with Piper's analysis. Moreover, the two first *v*' levels of N_2(C) state are mainly produced by the pooling reaction (see Fig. 3). The excitation transfer from Ar^m atoms to N_2 (reaction (R_4)) is an efficient channel only for N_2(C, *v*'=0) state. The Fig. 3 shows that the weight of (R_2) reaction, i.e. electron impact from N_2(A) specie, for the N_2(C, *v*' ≥ 2) state kinetics, becomes significant whatever the gas mixtures. The related rate coefficients, not known presently, are assumed to be roughly proportional to the FC factors. This fact could explain the discrepancies between experimental and theoretical vibrational distributions for these levels. More experimental data about N_2(A) state and electrons could help to improve this strongly consistent state-to-state model.

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References

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