Kinetics of the nitrogen pink afterglow in the presence of oxygen impurities

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The nitrogen afterglow is investigated by studying its behavior under the addition of small ammounts of oxygen, up to 1%, at a pressure p=8 Torr. It is shown the the emissions of $N_2(B)$ and $N_2^+(B)$ are strongly reduced under the presence of very little quantities of oxygen, even when oxygen is introduced directly in the post-discharge. It is argued this is essentially an outcome of the significant production of oxygen atoms in the afterglow, which subsequently destroy the vibrationally excited molecules $N_2(X,v)$. Consequently, the effectivness of the V-E energy transfers responsible for the appearance of the pink afterglow is notably decreased.

The nitrogen pink afterglow is characterized by a raise in the populations of many different species occurring downstream from the discharge, in a field-free region and after their diminution at a dark zone. Such peculiar behaviour was observed for radiative states such as $N_2(B,C)$ and $N_2^+(B)$ – the latter emission dominating the visible emission and naming the afterglow –, metastable atomic and molecular states, $N(^2D,^2P)$ and $N_2(A,a)$, as well as electrons and ground state ions $N_2^+(X)$ [1-4]. This puzzling phenomenon has been shown to be the result of an energy exchange from the vibrationally excited $N_2(X,v)$ dark states to the molecular nitrogen electronic states, at least to $N_2(A,a')$, in the so-called V-E energy transfers [5]. Although the broad picture seems relatively well established [6], the magnitude of the relevant rate coefficients and many details of the kinetics are still surrounded by some uncertainties. One possibility to gather additional information on the kinetics involved in the pink afterglow is to study its behavior in the presence of small impurities, such as oxygen, as O and O_2 quench efficiently the species driving the pink afterglow, namely $N_2(X,v)$ and $N_2(A,a')$. The purpose of the present work is to investigate the nitrogen pink afterglow by following this strategy.

A series of optical emission spectroscopy measurements were performed in an RF discharge in the range 1-20 Torr, revealing a strong influence of a very small oxygen addition, up to 1%, and an efficient destruction of the pink afterglow. Oxygen was added either in the discharge or in the afterglow, the former having a more pronounced effect than the latter. Herein we report the measurements made at p=8 Torr, when oxygen is added in the afterglow.

We have analysed the experimental results with the help of a self-consistent kinetic model, describing a N_2-O_2 stationary discharge and its afterglow. The model takes into account the electron kinetics, described by the homogenous electron Boltzmann equation solved under the two-term expansion in spherical harmonics, which is coupled to a system of rate balance equations describing the heavy-particle kinetics, including the complete manifold of vibrational levels of ground-state $N_2(X)$ molecules, several molecular N_2 and O_2 electronic excited states, ground-state and excited N and O atoms, as well as various radicals and ions. The dischare was considered to take place in a cylindrical tube with radius R=0.3 cm and the gas temperature to be T_g=500 K, whereas these values were changed to 1.05 cm and 300 K, respectively, in the afterglow.

Modelling results confirm the same trend as the experiments. In the first instants of the afterglow O atoms are formed in collisions involving N_2 electronically excited states, mainly $N_2(B)$, and later via the reaction N+NO→N_2+O. Oxygen atoms will shape the vibration distribution function of $N_2(X,v)$ molecules, in V-T collisions and by the Zel'dovich NO formation reaction, $N_2(X,v\geq13)$+O→NO+N. The rate coefficients of both mechanisms are poorly known. In the present calculations we have kept the rate coefficient for V-T deactivation with O atoms used in previous works [7] and tested two values for the Zel'dovich reaction, k=10^{-13} cm³/s [7] and 10^{-11} cm³/s [8], with no v-level dependence, as shown in Figure 1. A good agreement between the model calculations and experimental results was obtained in the latter case, confirming that the global rate of deactivation of $N_2(X,v)$ molecules by O

atoms is very high. Work is in progress to perform calculations of both rates. Additional experimental results on this system can be found in a companion contribution to this conference [9].
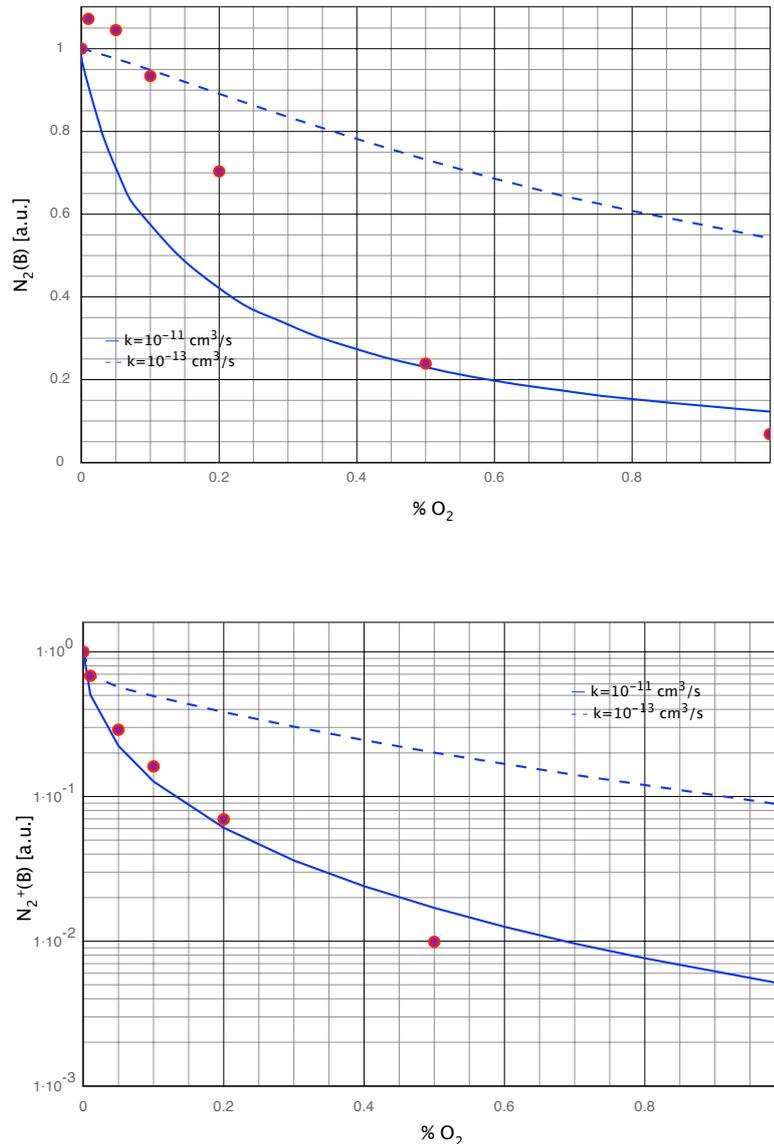

Fig. 1: Measured (points) and calculated (curves) variation of the population of $N_2(B)$ and $N_2^+(B)$ states, as a function of the O_2 concentration added into the post-discharge.

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