MODELLING OF AN ICP DISCHARGE IN OXYGEN WITH FULL KINETICS SCHEME WITH NEWLY CALCULATED VV/VT RATE CONSTANTS

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Abstract. In this work, 2D model of inductively coupled plasma discharge in oxygen were developed. The complete scheme of the vibrational kinetics of O_2 molecules was included in the model. The rate constants of the excitation and de-excitation processes for vibrationally excited levels of the O_2 molecule in reactions with O atoms were calculated based on obtained inelastic cross-sections of the O_2 + O system in the ground state for a set of initial vibrational and rotational states of O_2 molecule. These rate constants as well as calculated vibrational electron scattering cross-sections of O_2 were included in model and the results were compared with literature data. The vibrational distribution function was calculated in low-pressure inductively coupled discharge. Spatial distributions of plasma parameters were also obtained.

1. INTRODUCTION

Non-equilibrium low-temperature oxygen plasma has a wide range of applications. The use of such plasma has literally revolutionized many industrial processes such as plasma etching, surface treatment, plasma sterilization and medicine. The study of vibrational excitation and relaxation in oxygen plasma is actively studied area (see, for example, Andrienko 2020) and the experimental methods for detecting the vibrational distribution function in oxygen are rather complex. New experimental data on the distribution of vibrational excitation in stationary and nonstationary discharges in oxygen was recently obtained, see Annušová et al. 2018, and this indicates the need to develop more accurate models to describe the relaxation

kinetics of vibrational excitation under conditions of significant gas dissociation and to adapt these processes into complete self-consistent models.

2.   CALCULATION OF VV/VT CONSTANTS

The rate constants of the excitation and de-excitation processes for vibrationally excited levels of the O_2 molecule in reactions with O atoms were calculated based on obtained inelastic cross-sections of the $O_2 + O$ system in the ground state for a set of 8 initial vibrational and 256 rotational states of O_2 molecule, see Palov et al. 2024. Calculations of the vibrational energy exchange rate constants between two O_2 molecules were performed as well. The semi-classical coupled state method (see for example Hong et al., 2021, Kurnosov et al., 2023) was used in these calculations. The potential energy surface for the pair O_2-O_2 was set using an improved Lennard-Jones potential model formulated in Hong et al., 2021. Normalized cross-sections of vibrational excitation of O_2 molecule by electron impact were also obtained and included in our kinetic scheme. Overall, there were included 8 newly calculated first vibrational levels in addition to 25 highest ones taken from Annušová et al. 2018 of O_2 molecule for O_2+O collisional system with corresponding electron excitation cross-sections and VV/VT reactions in the ICP discharge kinetic scheme.

3.   ICP MODEL

The two-dimensional model used in this work has been described in detail elsewhere, see Kropotkin et al. 2019. Briefly, the model includes hydrodynamic equations in drift-diffusion approximation for describing the kinetics of charged (electrons and ions) and neutral plasma particles, Maxwell's equations for electromagnetic fields, and equations for the temperature and neutral gas flow as well as a detailed scheme of plasma-chemical reactions. The full scheme is available in Kropotkin et al. 2023. Before calculations for obtaining vibrational distribution function (VDF) in oxygen, the model was tested against theoretical and experimental data in a simple cylindrical chamber. The electron density and temperature distributions were obtained over a wide range of powers deposited in the plasma (100 – 500 W) and compared with literature data, see Kiehlbauch et al. 2003. The cylindrical discharge chamber was 16.2 cm in radius and 10.4 cm in height. The antenna was a flat four-turn coil separated from the discharge by a 1.5 cm thick quartz glass. The inductive discharge was simulated under the following conditions: the frequency was 13.56 MHz, the gas pressure was 10 mTorr, the power applied to the coils was from 100 to 500 W, and the gas flow was 33.5 sccm. Part of the calculation results is shown in Fig. 1.
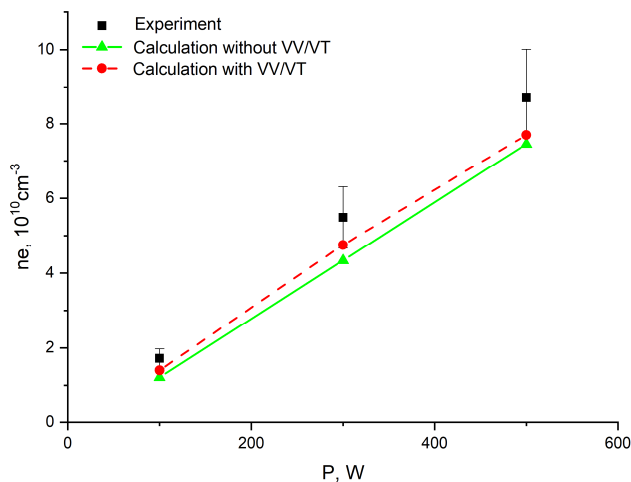
Figure 1: Electron concentration depending on the discharge power, calculated according to a simplified kinetic scheme and taking into account vibrational kinetics, in comparison with the experiment.

After verification of the model on experimental data the next goal was to compare VV/VT rate constants calculated during this work and available in literature, see Coletti et al. 2002, Esposito et al. 2008, Laporta et al. 2013, Lino da Silva et al. 2012. Calculations were carried out with an updated set of rate constants for VV/VT processes with O_2-O_2 molecules, and VT relaxation on O atoms. As can be seen from Fig. 1, the electron concentration remained practically unchanged (red dotted line), the change in values does not exceed 10%. Thus, the addition of a full set of vibrational kinetics did not affect the description of the main plasma characteristics by the model. The main focus of this work was on VDF in ICP discharge conditions. Carrying out such calculations taking into account a new set of excitation and relaxation rate constants will make it possible to clarify the form of the VDF in low pressure ICP plasma and DC glow discharge conditions (future work), and may also raise new questions about new resonant processes involving electronically excited states to explain the peculiarities of the VDF behavior observed in the experiments.

Acknowledgments

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