Argon metastable atom quenching in low pressure Ar/O_2 CCPs

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The kinetics of excited particles could exert a significant effect on low-temperature discharges. For CCPs operated in argon, stepwise ionization and pooling ionization involving argon metastable atoms provide alternative pathways beside direct ionization from the ground state by electron impact. Though the density of metastable levels ($1s_5$ and $1s_3$) is a few magnitudes lower than that of the ground state atoms, the lower energy thresholds of stepwise processes can lead to substantial reaction rates. It is indicated that the ionization from argon metastable atoms can surpass the direct one as the pressure approaches 1 Torr [1]. Nevertheless, the density of metastable atoms can decrease rapidly by a small amount of oxygen as shown by measurements [2] as well as numerical simulations [3]. The limited quantity of oxygen might come not only from intentional mixture but also as gas impurity in experiments. Hence one-dimensional kinetic simulations and experiments in argon/oxygen CCPs under low pressures are conducted to investigate the physical processes of argon metastable population and de-population.

Table 1: device parameters and discharge conditions.

Physical parameter	**Value**
Electrode gap	4 cm
Electrode diameter	14 cm
Driving frequency	13.56 MHz
Peak-to peak voltage	60-300 V
Gas pressure	2-10 Pa
Oxygen ratio	0-6%

The experimental setup consists of a geometrically symmetric cylindrical discharge chamber and tunable diode laser absorption spectroscopy (TDLAS) system for argon metastable atom density measurements. Some of the device parameters and discharge conditions are shown in Table 1. The transitions having wavelengths 772.376 nm and 772.421 nm are employed in TDLAS for $1s_5$ and $1s_3$, respectively.

The simulation model for argon/oxygen plasmas is based on a full model [4] for pure argon discharges. Simplification has been made to reduce computation time while preserving good agreement at low pressures, which is proved by code-to-code benchmarks in pure argon cases. The simplified model consists of basic 1d PIC/MCC code and extended diffusion-reaction-radiation (DRR) code. The former includes the charged particles and the oxygen metastable $O_2(a^1\Delta g)$ due to its important contribution to quenching of negative oxygen ions. The electron impact excitation rates from ground state (GS) argon atoms to 30 excited levels are recorded after the convergence of the PIC code and are used as inputs for the DRR code. Taken the excitation rates as sources, the diffusion equations of the two metastable levels are solved in the DRR code to get their density distributions,

$$-D\frac{\partial^2 n_m}{\partial x^2} = S_{\mathrm{GS}} + S_{\mathrm{rad}} + S_{\mathrm{cas}} - L_{\mathrm{rad}} - L_{\mathrm{step}} - L_{\mathrm{oxy}}, \quad (1)$$

where D is the diffusion coefficient, n_m refers to the metastable density. "S" represents the source of the Ar metastable levels from GS excitation, radiation from 2p levels and cascades from Rydberg levels with subscripts of "GS", "rad" and "cas", respectively. Similarly, "L" with different subscripts stands for the losses due to radial diffusion, stepwise excitation of Ar atoms, and quenching of Ar metastables by O_2 molecules with a rate coefficient taken from [5]. Eq. (1) is solved with a boundary condition taken from [6].

The computed and experimental densities at the centre of electrode gap are presented here. For pure argon, the metastable densities always increase initially with the rise of discharge voltages and tend to saturate as shown in Fig. 1. The ratios of the computed densities to the experimental values are approximately 2-3, which is reasonable considering the performance of the full model and experimental uncertainties.

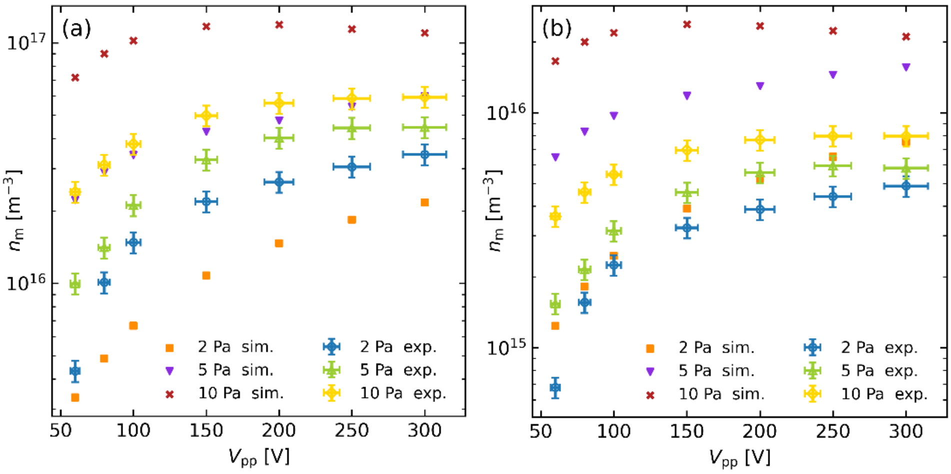

Fig. 1: Densities of Ar metastable atoms: (a) $1s_5$ and (b) $1s_3$, at the centre of electrode gap spacing at different pressures as a function of RF peak-to-peak voltage.

With the addition of oxygen, the metastable densities decrease rapidly, and then, the trend slows down when the gas pressure and discharge voltage are fixed. Good agreement between the simulations and the experiments is achieved for both metastable levels. The results prove that the simplified model is applicable for argon/oxygen CCPs at low pressures.

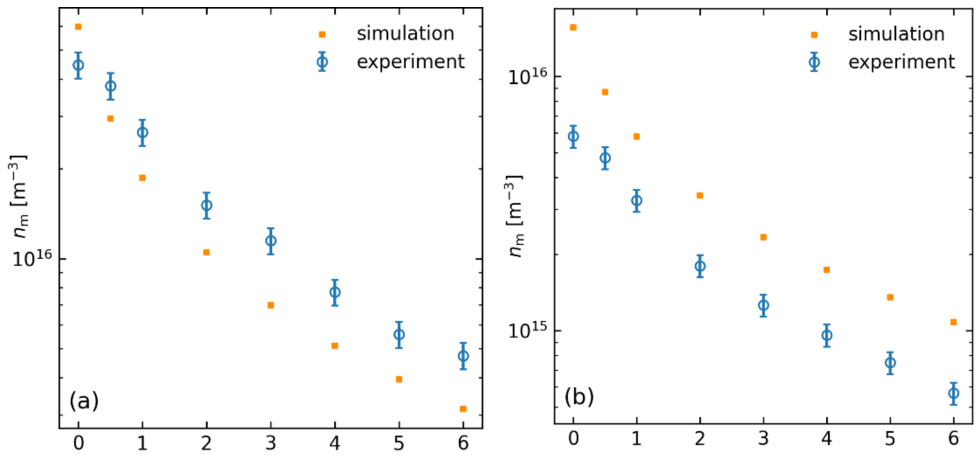

Fig. 2: Densities of Ar metastable atoms: (a) $1s_5$ and (b) $1s_3$ at the centre of electrode gap at 5 Pa and 300 V, as a function of oxygen content in the gas mixture.

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