Electron bounce resonance heating in dual-frequency capacitive coupled oxygen discharges

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Abstract: The BRH in DF CCPs operated in oxygen has been studied by different experimental methods and a particle-in-cell/Monte Carlo collision (PIC/MCC) simulation, and compared with the electropositive argon discharge. All these experimental observations are explained by PIC/MCC simulations, which show that in the oxygen discharge the bulk electric field becomes quite strong and is out of phase with the sheath field.

Keywords: Dual-frequency, electron density, hairpin probe, oxygen discharge

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1. Introduction

Recently, the bounce resonance heating (BRH) effect has been experimentally observed in DF CCPs in electropositive argon, and it was found to markedly enhance the discharge performance under certain conditions. However, in real applications, electronegative gases are most frequently employed. They are different from electropositive gas discharges in some basic, important features, such as the development of double layers [1, 2], a strong electric field and thus a high ionization rate in the bulk [3], more pronounced modulation of the bulk electric field [4, 5], and a discharge mode transition induced by voltage or pressure[6-8]. These pronounced differences will have a substantial impact on the BRH, and the resultant electron energy probability function (EEPF) determines the basic discharge dynamics, such as dissociation, excitation, and ionization in electronegative discharges. Therefore, it is of fundamental importance to study the features of BRH in electronegative discharges.

2. Experiment setup and simulation

Our experiments are performed in a parallel-plate DF (2/60 MHz) capacitive discharge chamber [9]. When conducting the optical measurements, the O2 discharge is diluted with 5% Ar, as it is difficult to probe the emission from the direct excitation of molecule O2. The measured electron density by the hairpin probe is corrected by using the fluid model described by Piejak [10]. In the experiment, we will compare the different behavior of BRH in Ar and O2 and study the effect of high-frequency (HF) power on the BRH in the O2 discharge.

Fig. 1 Schematic diagrams of the dual-frequency capacitively coupled plasma reactor.

Our simulations, which serve as a numerical explanation of the experiments of O2 and Ar discharges,
are based on the standard one-dimensional in space three-dimensional in velocity (1D3V) electrostatic PIC MCC method. More details can be found in Ref. [9]. To shed light on the BRH behavior at different HF powers in the experiments of O2, we compare the spatiotemporal distribution of various plasma quantities (i.e., the electron impact ionization rate, bulk electric field, trajectory of a typical bouncing electron and its energy) for a range of HF voltages. In the case of the O2 discharge, the charged particles O2+, O, and the electrons are traced using cross sections from Ref. 23. For Ar, the cross sections are the same as in Ref. [9].

3. Results and discussion

3.1 electrode gap effect

Fig. 2 Experimental measurements: positive ion and electron density versus gap length, in Ar (black) and O2 (red lines) discharges, at a fixed HF and LF power of 30W and 100 W, respectively. Note that the pressure increases from 1.3 Pa in Ar to 4.0 Pa to sustain the O2 discharge.

Fig. 2 illustrates the measured positive ion density and electron density versus gap length L, at a fixed HF and low-frequency (LF) power of 30W and 100 W, for the Ar and O2 discharge (black and red curves, respectively). The Ar+ ion density is measured in absolute terms, whereas the O2+ density is only obtained in relative terms. The BRH effect is illustrated by the peak in ion and electron density at a certain gap length, called the resonant gap length. From this figure, the differences of the BRH effect between Ar and O2 discharges can be generally summarized as followings. (i) Compared with the Ar discharge, the most significant BRH effect in the O2 discharge occurs at a somewhat larger gap, with the resonant peak broadened. The larger resonance gap and broader peak in the O2 discharge is related to the electronegative discharge structure. In simulations, we observed a narrower bulk region and higher bulk electric field. Note that the position of the resonance peak is almost independent of the pressure in both Ar [9] and O2.

(ii) At L > 2.5 cm, the positive ion and electron densities drop monotonously in the O2 discharge upon increasing L, while the plasma density rises in the Ar case. This is due to the more significant drop of the BRH with increasing L in the O2 discharge. It is worth to mention that our PIC MCC simulations have qualitatively reproduced the experimental density curves both in Ar and O2.

3.2 HF power effect

Fig. 3 Experimental measurements in the O2 plasma: relative O2+ ion density at the discharge center versus L, at a range of HF powers PH for a fixed LF power PL of 100 W, at a gas pressure of 4.0 Pa.

The effect of HF power PH on the BRH in the O2 discharge is shown in Fig. 3, which displays the measured relative O2+ ion density versus L at different values of PH with a fixed PL of 100W and a pressure of 4.0 Pa. At lower PH, both the O2+ ion density peak gradually drops and almost disappears at PH = 20 W, which indicates a suppressed BRH. It should be noted that the results are quite different from the electropositive Ar discharge, where the BRH is hardly affected by PH [9]. This is attributed to the fact that the O2 discharge experiences a mode transition from electropositive to
electronegative due to the lower amount of sheath heating with the drop of PH. This mode transition induced by the driving voltage has been observed in CCPs with other electronegative gases as well [6, 7].

3.3 Gas pressure

Fig. 4 illustrates the measured positive-ion O2+ density versus electrode gap L at different pressures. At higher pressures, the maximum around the resonance gap diminishes gradually and almost disappears at 12 Pa, which indicates again a suppression of the BRH. In electronegative discharges, two different pressure effects are responsible for the BRH. First, as the electron mean free path is inversely proportional to the pressure, the resonance electrons simply cannot bounce long time enough at high pressures, to gain sufficient energy before being stopped or re-directed by a collision. This effect also occurs in a pure argon discharge [12]. Moreover, an additional pressure effect occurs in the oxygen discharge, namely the enhanced bulk electric field reversal, which suppresses the BRH at higher pressures.

![Graph](image)

Fig. 4 Measured positive-ion (O2+) density versus electrode gap L in the oxygen discharge at different pressures with a fixed HF and LF power of 30W and 100W

3.4 HF frequency effect

Fig. 5 illustrates the phase delay of the plasma bulk field with respect to the sheath field, as a function of the driving frequency. At the driving frequency of 13.56 MHz, the bulk electric field is in phase with the sheath, so θ is zero. With the increase in the driving frequency, the phase of the bulk electric field will delay with respect to the sheath field, the phase delay θ increases rapidly and tends to reach its maximum π/2 when the driving frequency approaches 120 MHz. This behavior can be explained because at relatively low driving frequency the electrons can respond to the rf electric field whereas they gradually fail to follow the rf field at higher frequencies, i.e. at the driving frequency f > 60 MHz. Note that this phase relationship between the plasma bulk field and the sheath field at different driving frequencies is consistent with the results in [13].

![Graph](image)

Fig. 5 Phase delay of the plasma bulk field with respect to the sheath field, θ, as a function of the driving frequency.

4. Conclusion

We investigated the electron bounce resonance heating (BRH) in dual-frequency capacitively coupled plasmas operating in oxygen by different experimental methods and a particle-in-cell/Monte Carlo collision (PIC/MCC) simulation, and compared with the electropositive argon discharge. In comparison with the electropositive argon discharge, the BRH in the electronegative oxygen discharge tends to happen at larger electrode gaps. This is related to the peculiar oxygen discharge structure, i.e. the development of a double layer in the sheath and the narrower bulk region and higher reversal electric field in the bulk. At electrode gap L > 2.5 cm, the positive-ion (and electron) density drops monotonically in the oxygen discharge upon increasing electrode gap, whereas it rises (after an initial drop) in the argon case. This indicates that the BRH drops significantly at electrode gaps larger than the resonance length, in the oxygen discharge. PIC/MCC simulation results show that in the oxygen discharge, the bulk electric field reversal becomes quite
strong and is out of phase with the sheath field. Therefore, it retards the resonance electrons when traversing the bulk, resulting in a suppressed BRH. Finally, in addition to the effect of the discharge gas (oxygen versus argon), also the effect of HF power, gas pressure and high frequency on the BRH were investigated, both experimentally and by simulations. It appears that the resonance peaks in the positive-ion density become weaker with decreasing HF power. This is attributed to the enhancement of the reversal bulk electric field when the discharge undergoes a transition from electropositive to electronegative mode with the drop in the HF power. Furthermore, in the pure oxygen discharge, the BRH is suppressed with increasing pressure and it almost diminishes at 12 Pa. This is due to a combined effect of more frequent electron–neutral collisions and enhanced bulk electric field reversal at high pressures. Finally, the BRH was found to depend significantly on the high frequency applied, which can be explained based on the phase relation between bulk electric field and sheath electric field.

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Reference