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Modes of low-pressure dual-frequency (27/2 MHz) discharges in hydrogen

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Abstract
This paper studies the modes of dual-frequency (high-frequency (HF)/low-frequency (LF)) low-pressure discharges. The dual-frequency discharges are shown to burn in one of three possible modes. At small LF voltages the first mode is observed, i.e. the HF discharge perturbed by the LF voltage. The second mode, i.e. the combined discharge, exists in the presence of intense ionization in the sheaths, when the LF voltage exceeds some critical value. The third mode (the LF discharge perturbed by an HF field) is observed when a small HF voltage is applied to the burning LF discharge. The range of parameters within which the first mode of the combined discharge may be extinguished by the LF voltage increase is shown to be limited by the HF discharge extinction curve from the low-pressure side as well as the lowest HF voltage for the transition of the discharge from the first mode to the second one.

1. Introduction

Dual-frequency discharges recently found broad application in the technological processes of etching SiO_2 [1–5], Si and Si_3N_4 [4], SiOC [6], platinum [7, 8], ruthenium [9], depositing SiO_2 [10], Si_3N_4 [11], fluorinated silicon dioxide films [12] and of plasma surface modification of hydrogel thin films [13].

A conventional capacitive RF discharge burning at a fixed frequency f in a discharge chamber with an inter-electrode gap L, gas pressure p and a definite value of the RF power P_{dlv} keeps the ion flow to the electrodes and the ion energy fixed. When one changes, e.g. the gas pressure or the magnitude of the RF power, then the ion flow to the electrodes together with the ion energy would change simultaneously. Therefore separately controlling these two quantities in a usual single-frequency discharge is impossible. However, in a number of technological processes using an RF discharge it is necessary to control the values of the ion density and the energy of ions bombarding the electrodes (materials and films under processing) independently. A high concentration of plasma ensures a high dissociation degree of chemically active gases enhancing the film deposition rate as well as the etching rate of semiconductor materials. Increasing the ion energy increases the rates of sputtering and etching films. The problem of separately controlling the ion flow to the electrodes and the ion energy is solved easily in an RF discharge with two frequencies because a high-frequency (HF) of the electric field provides a high ionization rate of gas molecules via electron impact and a dense plasma, whereas a low-frequency (LF) increases the voltage drop across the near-electrode sheath and, as a consequence, accelerates ions to higher energy.

Therefore increased interest in the experimental and analytical study of the processes occurring in dual-frequency discharges was observed recently. Jaiprakash and Thompson [14] examined 100 kHz and 12.2 MHz discharges and found that electron concentrations increase with increasing total power and decrease with increasing levels of LF power. Vahedi and DiPeso [15] used a particle-in-cell code to simulate a dually excited capacitively coupled RF discharge and showed how such a system can be used as a plasma processing tool with a separate control over ion flux and ion bombarding energy. Kitajima *et al* [16] investigated two-dimensional images of a dual-frequency (13.56 MHz and 100 MHz/700 kHz) capacitively coupled plasma by using 2D-t optical emission spectroscopy. Makabe and Maeshige [17] summarized models of low-temperature plasma, considering also the model of dual-frequency discharges. Robiche *et al* [18] proposed an analytical sheath model for the RF discharge operated with two frequencies and studied under the assumption of time-independent, collisionless ion motion. Franklin [19] simplified the expressions for the time-average electric potential within the sheath and the sheath boundary motion, obtained in [18]. Wakayama and Nanbu [20] examined the dynamic structure of dual frequency using self-consistent particle-in-cell/Monte Carlo (PIC/MC) simulation. Kim *et al* [21] analysed a homogeneous plasma model for dual radio-frequency discharges driven by two sinusoidal current sources and obtained analytical expressions for discharge parameters as a function of the effective frequency, effective current and effective voltage. Boyle *et al* used the PIC code to show that the most important governing parameter is the ratio of the driving frequencies [22] and investigated the nonlinearity of the plasma sheath in dual-frequency rf discharges for frequencies above the ion plasma frequency and for the collisionality regime [23]. Georgieva *et al* [24] numerically investigated ion-energy-distribution functions (IEDFs) in rf single and dual frequency discharges by a one-dimensional PIC/MC model. Lee *et al* [25] used PIC/MC simulation to study the possibility of IEDF control in asymmetric single and dual frequency discharges. Turner and Chabert [26] discussed electron heating mechanisms in the sheath regions of dual-frequency capacitive discharges and showed that the heating effect produced by either Ohmic or collisionless heating is much larger when the discharge is excited by a superposition of currents at two frequencies than if either current had acted alone.

As we see, recently a large number of experimental and theoretical papers appeared which were devoted to studying various characteristics of dual-frequency discharges. However, as far as we know, there is still a lack of papers describing different modes of such discharges related to different values of HF and LF voltages.

We studied dual-frequency (HF/LF) discharges in the experiment. We applied the HF voltage and additionally the LF voltage U_{lf} simultaneously across the electrodes. As a result, we found that dual-frequency discharges might exist in three modes. At low HF voltages the additional LF voltage makes a small contribution to the ionization rate of gas molecules via electron impact within the plasma volume, and we observe the first mode of burning of the dual-frequency discharge—a 'non-self-sustained HF discharge perturbed by the LF voltage'. When the HF voltage and the LF voltage are sufficiently large to induce the breakdown of the sheaths, the discharge experiences the transition to the second mode—a 'combined discharge'. Only this mode is of considerable interest for plasma technology. The third mode may be obtained applying a small HF voltage to the burning LF discharge. We will call it conditionally a 'non-self-sustained LF discharge perturbed by the HF voltage'.

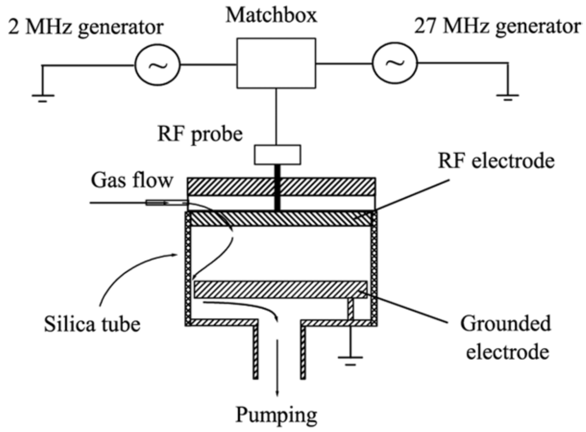

Figure 1. Set-up of the experimental device.

2. Experimental

The RF discharge was ignited in H_2 over the pressure range $p \approx 0.01$–10 Torr with RF field frequencies of $f_{lf} = 2$ MHz (LF) and $f_{hf} = 27.12$ MHz (HF). The distance between the flat circular aluminium electrodes (143 mm in diameter) was equal to $L = 20.4$ mm. The HF and LF voltages (amplitude <1500 V) were applied to one electrode, while the other was grounded. The electrodes were located inside a fused silica tube with an inner diameter of 145 mm (see figure 1). The gas was supplied through small orifices in the powered electrode and then pumped out via the gap between the second electrode and the wall of the fused silica tube.

The gas pressure was monitored with 10 and 1000 Torr capacitive manometers (MKS Instruments). The gas flow was fixed with a mass flow controller to 5 sccm, and the pressure regulated by throttling the outlet to the pump. The rf voltage was measured with an RF current–voltage probe (Advanced Energy Z'SCAN). This rf probe was located at the minimum possible distance from the RF electrode. This probe gave measurements of the rf voltage, rf current, phase shift angle φ between current and voltage and delivered power for both HF and LF frequencies.

3. Experimental results

We are going to study the effect of the LF voltage $U_{2\,\mathrm{MHz}}$ on the properties of the HF discharge with the frequency of 27.12 MHz. Therefore it is expedient to first consider the characteristics of the unperturbed HF discharge. Figure 2 shows the current–voltage characteristics (CVCs) of the HF discharge at different values of hydrogen pressure. It is clear from the figure that at a low pressure of $p < 0.1$ Torr the HF Ohmic current $I_{rf}\cos\varphi$ first grows uniformly with the HF voltage $U_{27\,\mathrm{MHz}}$ increasing. However after the quantity $U_{27\,\mathrm{MHz}}$ approaches some critical value, the discharge current decreases abruptly, and then the discharge is extinguished. The higher the hydrogen pressure, the higher the HF voltage $U_{27\,\mathrm{MHz}}$ at which the discharge is extinguished. The reasons for such a phenomenon can be understood considering the extinction curve of the HF discharge $U_{27.\mathrm{ext}}$ depicted in figure 3. In the

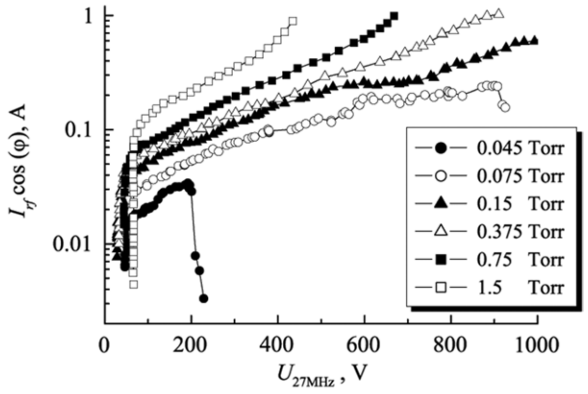

Figure 2. CVCs of the HF discharge with the frequency of 27.12 MHz for various hydrogen pressure values.

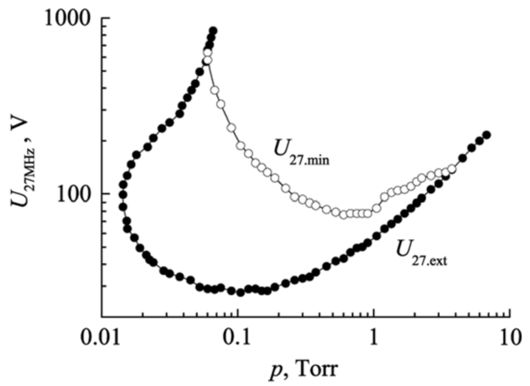

Figure 3. HF extinction voltage $U_{27.\mathrm{ext}}$ of the self-sustained HF discharge ($f = 27.12$ MHz) and the smallest HF voltage $U_{27.\mathrm{min}}$ of the transition from the first mode of the dual-frequency discharge to the second one against hydrogen pressure.

pressure range of $p < 0.1$ Torr this curve possesses a region where the dependence of the HF extinguishing voltage on the gas pressure is multi-valued. That is, in this pressure range the HF discharge may be extinguished not only through decreasing but also through increasing the HF voltage $U_{27\,\mathrm{MHz}}$. Besides, a minimum hydrogen pressure value exists, $p = 0.014$ Torr, at which the HF discharge sustainment is possible. At lower pressure values the HF discharge cannot exist with this inter-electrode gap value (within the range of HF voltage values that we studied). This phenomenon was already studied in [27,28], therefore we will not pay attention to it here.

At pressure values of $p > 0.1$ Torr increasing the HF voltage does not lead to the HF discharge extinction. With the HF voltage growing the discharge current increases uniformly, and the discharge experiences a transition from the α to the γ mode [28–38]. At pressure values of $p \geqslant 0.4$ Torr the α mode exists in the normal regime before extinction, i.e. the decrease in the discharge current occurs with almost constant HF voltage due to the decrease in the area occupied by the plasma column on the electrode.

Let us compare the characteristics of the discharges at 2 and 27.12 MHz. Figure 4 shows the CVCs $I_{\mathrm{rf}}\cos\varphi$ of the discharges for these two frequency values with the fixed hydrogen pressure of $p = 0.75$ Torr. It is clear from the figure that high voltage values $U_{2\,\mathrm{MHz}} \geqslant 354$ V are required

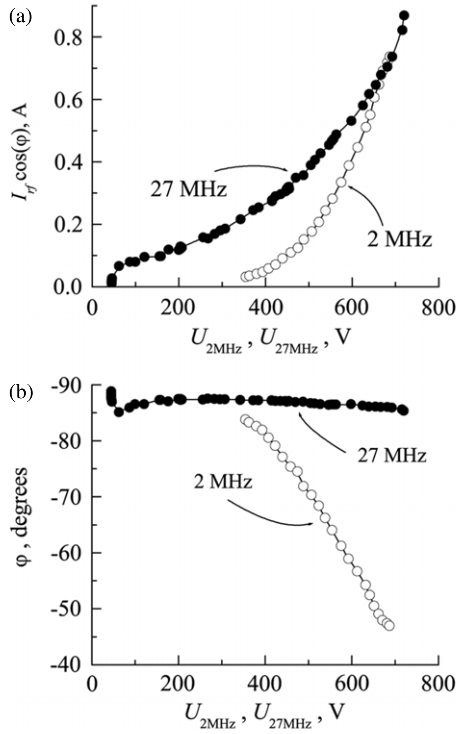

Figure 4. CVCs (*a*) of the HF and LF discharges as well as phase shift angles (*b*) for the frequencies of 27.12 and 2 MHz with hydrogen pressure of 0.75 Torr.

to sustain the LF discharge whereas the HF discharge may exist at $U_{27\,\mathrm{MHz}} \geqslant 45$ V. The CVC of the HF discharge before extinction contains a section with the normal regime. However we did not observe the normal regime in the LF discharge within the total range of hydrogen pressures that we studied.

Figure 4 also shows the phase shift angle φ between the current and the voltage for frequencies 2 and 27.12 MHz. The φ values for 27.12 MHz indicate the capacitive nature of the discharge at this frequency, and they are contained within the range $\varphi \approx -85^\circ$ to -89° in the total range of HF voltages $U_{27\,\mathrm{MHz}}$ studied. The discharge at $f = 2$ MHz is far more resistive, i.e. on increasing the LF voltage $U_{2\,\mathrm{MHz}}$ from 354 to 687 V the phase shift angle varies uniformly from -84° to -47°.

Let us now consider how the parameters of the HF discharge vary on applying the LF voltage $U_{2\,\mathrm{MHz}}$ across it. Let us ignite the HF discharge in the weak-current α-regime, in which the ionization occurs within the region of the quasi-neutral plasma, and let us apply a small LF voltage across the electrodes. The thickness of the sheaths near the electrodes d_{sh} increases (see figure 5). In this case we observe the first mode of the dual-frequency discharge—a *non-self-sustained HF discharge perturbed by the LF electric field*. If the HF voltage does not exceed the limiting value $U_{27.\mathrm{min}}$, a further increase in the LF voltage entails the decrease in the plasma region width, and at some $U_{2\,\mathrm{MHz}}$ value the discharge goes out. At the same time, the less the HF voltage value sustained, the less the LF voltage $U_{2\,\mathrm{MHz}}$ required for discharge extinction. However, on increasing $U_{2\,\mathrm{MHz}}$ further the joint action of LF and HF voltages leads to gas breakdown, and the discharge ignites in the second mode which we conditionally call a '*combined discharge*'.

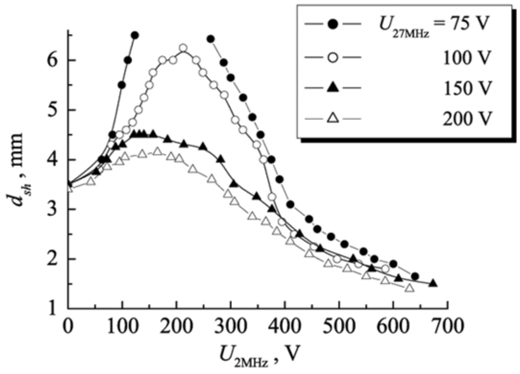

Figure 5. Sheath thickness against the LF voltage at different HF voltage values and hydrogen pressure of 0.75 Torr.

Figure 3 depicts the dependence of the critical HF voltage $U_{27.min}$ on gas pressure. At $U_{27\,MHz} = U_{27.min}$ the discharge with LF voltage growing experiences a transition to the second mode without going out. It is clear from figure 3 that at low gas pressure and high HF voltage the extinction curve of the HF discharge $U_{27.ext}$ and the curve of the critical HF transition voltage to the second mode $U_{27.min}$ merge. Therefore the ranges of pressures and HF voltages in which the first mode of the dual-frequency discharge can be extinguished through the application of the LF voltage are limited from the low-pressure side and are contained within the gap between curves $U_{27.ext}$ and $U_{27.min}$. It does not mean that the existence range of the first mode of the dual-frequency discharge is contained only within the gap between curves $U_{27.ext}$ and $U_{27.min}$. The first mode is also observed at HF voltages $U_{27\,MHz} > U_{27.min}$, but with the increase in the LF voltage the discharge experiences a transition to the second mode without going out. At the same time, if in the first mode the sheaths were dark and with the $U_{2\,MHz}$ increasing became broader, then after the transition to the second mode the luminosity of violet tint appeared in the sheaths (indicating the appearance of high-energy electrons in the sheath), and its thickness decreased with the growth in the LF voltage. Note also that at pressures above 3.8 Torr we failed to extinguish the first mode of the dual-frequency discharge on applying the LF voltage, the discharge after a certain increase in the sheath thickness experiencing a transition to the second mode. Therefore the ranges of pressures and HF voltages in which the first mode of the dual-frequency discharge can be extinguished through the application of the LF voltage are also limited from the high-pressure side.

Figure 6(*a*) shows the phase shift angle φ between the current and the voltage for frequencies of 2 and 27.12 MHz against the LF voltage $U_{2\,MHz}$ applied to the discharge. It is clear from the figure that in the first mode of the dual-frequency discharge with $U_{2\,MHz}$ growing the phase shift angle for the frequency 27.12 MHz φ_{27} experiences a weak variation (from $-86°$ to $-87.6°$), and the phase shift angle for the frequency 2 MHz φ_2 grows from $-85°$ to $-80°$. At $U_{2\,MHz} \approx 270$ V the dual-frequency discharge experiences a transition to the second mode. Electron avalanches develop in sheaths, the conductance of the sheaths increasing abruptly. It leads to a fast growth of the phase shift angle φ_2 with the LF voltage,

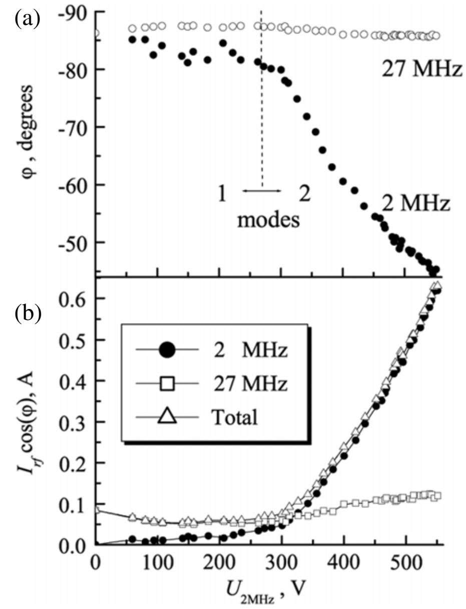

Figure 6. Phase shift angle for frequencies 27.12 and 2 MHz (*a*), Ohmic HF and LF currents and total current (*b*) of the discharge against the LF voltage at the hydrogen pressure of 0.75 Torr and HF voltage of $U_{27MHz} = 100$ V.

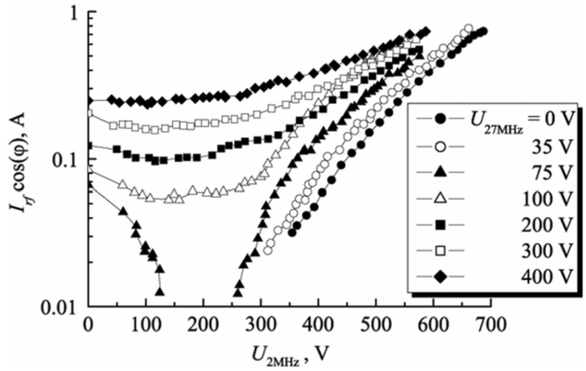

Figure 7. Total current against the LF voltage at the hydrogen pressure of 0.75 Torr and various HF voltage values.

up to $-45°$ at $U_{2\,MHz} \approx 550$ V. At the same time the phase shift angle φ_{27} increases slowly to $-85°$.

Figure 6(*b*) shows the Ohmic current ($I_{rf}\cos\varphi$) of the dual-frequency discharge for frequencies of 2 MHz (I_2) and 27.12 MHz (I_{27}), as well as the total discharge current $I_{total} = \sqrt{I_2^2 + I_{27}^2}$ against the LF voltage. On increasing the LF voltage the LF current I_2 first increases slowly and then the HF and the total current decrease. After the transition to the second mode (at $U_{2\,MHz} \approx 270$ V) a fast growth of the LF and the total current is observed.

Figure 7 depicts the total current against the LF voltage applied with the hydrogen pressure fixed at $p = 0.75$ Torr and different values of the HF voltage. At the small HF voltage (in the figure it corresponds to the curve for $U_{27\,MHz} = 35$ V) applied to the LF discharge we observe the third mode—a *non-self-sustained LF discharge perturbed by the HF electric field*. It is not possible to have a self-sustained HF discharge at these values of the HF voltage and hydrogen pressure.

However, even a small HF voltage affects the characteristics of the LF discharge noticeably. At a higher HF voltage the discharge experiences a transition from the third mode to the second one, but in this case it is difficult to choose a criterion for such a transition. Starting from $U_{27\,\mathrm{MHz}} = 50$ V, it is possible to sustain the HF discharge in the gap. Application of the LF voltage first gives rise to the first mode of the dual-frequency discharge. This mode at a low HF voltage is accompanied by the decrease in the total discharge current and then it goes out. At sufficiently high LF voltage a breakdown of the discharge gap occurs due to gas molecules' ionization via electron impact in the effective HF/LF field:

$$E_{\mathrm{eff}}^2 = \frac{\nu_{\mathrm{en}}^2}{\nu_{\mathrm{en}}^2 + \omega_{27\,\mathrm{MHz}}^2} \cdot \frac{E_{27\,\mathrm{MHz}}^2}{2} + \frac{\nu_{\mathrm{en}}^2}{\nu_{\mathrm{en}}^2 + \omega_{2\,\mathrm{MHz}}^2} \cdot \frac{E_{2\,\mathrm{MHz}}^2}{2}, \quad (1)$$

where ν_{en} is the electron–neutral collision rate, $\omega_{27\,\mathrm{MHz}}$ and $\omega_{2\,\mathrm{MHz}}$ are angular frequencies of the HF and LF fields and $E_{27\,\mathrm{MHz}}$ and $E_{2\,\mathrm{MHz}}$ are the strengths of the HF and LF electric fields, respectively. The dual-frequency discharge formed under these conditions exists in the second mode, and on increasing the LF voltage it is accompanied by the growth of the total current. At higher HF voltage values ($U_{27\,\mathrm{MHz}} \geqslant U_{27.\mathrm{min}}$ in figure 3) the dual-frequency discharge experiences a transition from the first mode to the second one without going out, the total current that decreased in the first mode increases fast with the LF voltage growing after the transition to the second mode.

Discharge extinction plays a role *only in the first mode* of the two-frequency discharge. Just in this mode at a low HF voltage, when the plasma concentration is small, increasing the LF voltage gives rise to the fast increase in the sheath thickness. This leads to the plasma volume narrowing, plasma density lowering and the decrease in the flow of positive ions out of the plasma to the electrodes. According to the Child–Langmuir law [30], the decrease of the ion flow through the sheath with a simultaneous increase in the LF voltage drop across the sheath is accompanied by the increase in the sheath thickness. When at a sufficiently large LF voltage the sheaths occupy a large part of the inter-electrode gap, the enhanced loss of ions out of the plasma ceases to be compensated by the ionization in the volume, and the discharge may go out. In the second mode the sheaths are broken, and electron avalanches develop in them. The flows of high-energy electrons leave the sheaths, thus leading to the increased ionization rate in the plasma volume and plasma concentration growth. Increasing the LF voltage involves only the amplification of electron avalanches within the sheaths, i.e. it makes discharge sustainment easier. In the third mode the sheaths are also broken, and the application of only a small HF voltage makes the development of electron avalanches easier, also it introduces a small contribution to the ionization of gas molecules via electrons having gained their energy in the effective electric field within the plasma volume. It is hardly possible to extinguish the second mode by increasing the LF voltage or the third mode by increasing the HF voltage.

Figure 8 shows the HF voltage values at which the transition from the first mode of the dual-frequency discharge to the second one occurs against the LF voltage applied with

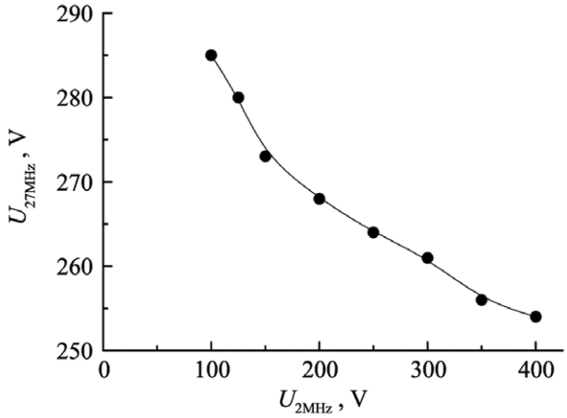

Figure 8. The HF voltage at which the transition from the first mode of the dual-frequency discharge to the second one occurs against the LF voltage applied at the hydrogen pressure of 0.75 Torr.

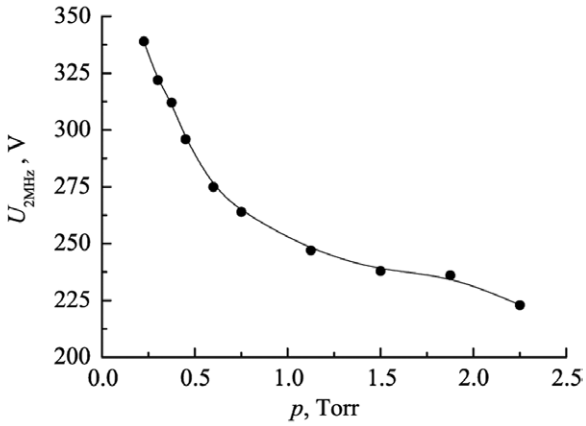

Figure 9. The LF voltage at which the transition from the first mode of the dual-frequency discharge to the second one occurs against hydrogen pressure and the HF voltage of 150 V.

the hydrogen pressure fixed. It is clear from the figure that the lower the HF voltage value, the higher the LF voltage required for the breakdown of sheaths and for the transition from the first mode of the dual-frequency discharge to the second one. It is due to the fact that not only practically all applied LF voltages are concentrated across the sheaths but also at certain moments of time almost all applied HF voltages 'drop' across them. It makes the sheath breakdown and the transition to the second mode considerably easier.

Figure 9 shows the LF voltage values at which the transition of the first mode of the dual-frequency discharge to the second one occurs against hydrogen pressure with the HF voltage fixed. The lower the gas pressure, the higher the LF voltage at which the transition from the first mode to the second one occurs. Perhaps it is associated with the fact that with decreasing pressure the thickness of the sheaths increases, and it is difficult to achieve the breakdown of thicker sheaths.

Here is the summary of the main properties of the three modes of the dual-frequency discharge.

(1) *Non-self-sustained HF discharge perturbed by the LF electric field.* Sheaths are not broken, there is no ionization in them. The main ionization, perhaps, occurs within the

plasma volume due to electrons having gained their energy in the effective RF electric field, as well as due to stochastic heating of electrons in the cathode phase of sheaths. Increasing LF voltage leads to the increase in the thickness of sheaths, the enhanced loss of positive ions from the plasma and, as a consequence, the discharge current decrease. This mode of the dual-frequency discharge may be extinguished within a certain range of HF voltages ($U_{27\,\mathrm{MHz}} < U_{27.\mathrm{min}}$) by the increase in the LF voltage, which in fact limits its application in plasma technologies.

(2) *Combined discharge.* Sheaths are broken, electron avalanches develop in them contributing to the ionization of gas molecules substantially. The HF electric field remaining sufficiently large supports the ionization in the plasma volume. The sheath thickness decreases with LF voltage growth due to the fast growth of the plasma concentration in the volume; the discharge current also grows.

(3) *Non-self-sustained LF discharge perturbed by the HF electric field.* The ionization in the sheaths in the LF electric field plays the main role in sustaining the dual-frequency discharge. The weak HF field makes a certain contribution to the gas ionization in the discharge volume. The sheath thickness decreases slightly with the HF voltage growing, and the discharge current grows.

4. Conclusions

We studied the modes of dual-frequency HF (27.12 MHz)/LF (2 MHz) discharges. At different ratios between HF and LF voltages three modes of the discharge are observed: (1) a non-self-sustained HF discharge, perturbed by an LF electric field, (2) a combined discharge and (3) a non-self-sustained LF discharge, perturbed by an HF field. The range of parameters within which the first mode of the dual-frequency discharge can be extinguished via increasing the LF voltage is limited on the low-pressure side by the HF discharge extinction curve as well as the curve of the smallest HF voltage corresponding to the transition of the dual-frequency discharge from the first mode to the second one. The conditions of existence of every mode of dual-frequency discharges are determined, and discharge CVCs are also recorded.

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