Azimuthal inhomogeneities of axially symmetric rf discharge plasma in arc-shaped magnetic field

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Abstract. An axially symmetrical rf magnetron discharge in argon with a small admixture of air and in air was studied. The plane loaded electrode was equipped with a ring-shaped dielectric insert. Design features of the electrode and of the magnetic system allow the discharge ring to be located in the region where the electric and magnetic field lines are not perpendicular to each other. It has been revealed that in this case the glowing plasma ring is sectionalized. Up to 10 long-living plasma bunches were observed. They equidistantly locate along the discharge ring and may either rotate around the discharge axis or remain motionless. Both the discharge power and the working gas pressure are quite different from those proper to dc and high-power impulse magnetron sputtering discharges with spokes.

1. Introduction

It was generally accepted several years ago that the plasma density in the magnetron discharge is homogeneously distributed across the region above the target racetrack. The development of high-speed camera technique made it possible to reveal the nonuniformities in form of plasma bunches (spokes, ionization zones) moving with a high velocity in the azimuthal direction. Such irregularities were discovered in high-power impulse magnetron sputtering (HiPIMS) [1–3] and dc magnetron discharges. As a rule, they follow one after another creating a periodic structure. Motion of nonuniformities in HiPIMS discharges usually coincides with the electron drift direction in the crossed $\mathbf{E} \times \mathbf{B}$ fields with the velocity one order of magnitude less than the electron $\mathbf{E} \times \mathbf{B}$ drift velocity [2]. Direction of movement reversal has been observed in varying the discharge current between dc and HiPIMS regimes [4]. In [5], two-dimensional periodical structures in a magnetized plasma on the condition of electric and magnetic field lines crossing at oblique angle are studied using kinetic simulations but the physics of this phenomenon has not been clarified.

Presently there are no established notions about the origination of plasma nonuniformities. They were discovered for the first time in [1] and it was pointed there that an irregular structure was energy-optimal for high current transfer. In [6] the reason for a plasma bunch formation was qualitatively explained by a positive feedback between stochastic growth of plasma density in some region of a racetrack and ionization probability by drift electron entering the same area. A plasma bunch formed in the area is characterized as a zone of enhanced potential and high electron temperature. According to this qualitative model, the electrons heated in discharge
presheath make most contribution to the bunch formation in comparison with the electrons emitted by the cathode. In [7], for a low current discharge, the origination of nonuniformities is connected with the gas breakdown, appearance of region with enhanced ionization and its subsequent movement as plasma bunch movement in the electron drift direction.

With appearance of plasma nonuniformity in axially symmetric magnetron discharge the azimuth electron drift in crossed $\mathbf{E} \times \mathbf{B}$ fields results in charge separation on its border and a double layer formation. Connected with a layer the electric field acts as an additional channel for heating plasma electrons. Moreover, together with magnetic field it leads to the electron drift perpendicular to the cathode surface [3, 8].

By now, azimuthal nonuniformities have been discovered in both unipolar pulsed (HiPIMS) and dc magnetron discharges where the secondary ion-electron emission plays a significant role. Magnetron discharge activation by the rf electric field expands the range of its application. Effective sputtering of dielectric materials by the rf magnetron discharge became possible. Irrespective to the presence of a magnetic field in the discharge area, in the capacitive rf discharge with flat electrodes there is a lack of conditions for development of instabilities along the discharge electric field [9]. Therefore rf discharges are used for active laser medium generation [9], for dusty plasma crystal formation and investigation [10]. The given magnetic field plasma of the rf discharge is capable to retain dusty particles and to deposit coatings on them [11–13].

In the present paper, the discharge with an unusual design of the electrode system was investigated. At first such a system was designated for dusty trap formation and dielectric coatings deposition on micron size particles. But it was found out that some features of the rf discharge are of separate interest. Namely, it is demonstrated that a plasma ring of the rf discharge in the presence of a magnetic field at specified conditions is uniformly segmented into bunches moving in the azimuth direction. The objective of the work is to study the relationship between the presence, form, quantity and velocity of azimuth irregularities on the one hand and the rf magnetron discharge parameters (power, self-bias) on the other hand.

2. Experiment
In the present work, the pattern of the rf discharge glow over a flat axially symmetric metal electrode (target) with axially symmetric dielectric cover was investigated. An axially-symmetric magnetic system was placed under the electrode. The radial cross-section of the electrode with cover and of the magnetic system is presented in figure 1(c). The experiments were carried out in argon, atmospheric air and a mixture of 99 vol % argon and 1 vol % air at pressure 1–13 Pa. The discharge was running at one of several available frequencies. At frequency 13.56 MHz the generator RF-VII RF-6-XIII with a matching system was employed. Low power returnable rf generator was used at frequencies 5.4, 6.4, 9.2 MHz. Direct power of the discharge was in 1–12 W range. Video camera with the recording rate 200 s$^{-1}$ was used for discharge observation. Transversal and longitudinal components of the magnetic field induction near the electrode surface were measured without plasma with a Hall sensor.

3. Results and discussion
Radial cross-sections of some different electrode systems and of the corresponding glow discharge regions are shown in figure 1. The center of the reference frame is placed in the middle of the electrode surface.

Over the electrode the magnetic field lines form two axially-symmetric arched systems. Internal and external arched systems are situated over $r_1$ and $r_2$ coordinates, respectively. In our experiments $r_1 = 2.5$ cm, $r_2 = 9$ cm. For a sufficiently thick target the magnetic induction on the electrode surface is in the range 0.01–0.08 T that is optimal for magnetron discharge burning. In the condition the plasma concentrates near the electrode in two ring regions where magnetic field lines are parallel to the electrode surface close to $r_1$ and $r_2$ coordinates. Usually
Figure 1. Diametral cross-sections of glow discharge region, the electrode and magnetic systems and magnetic field lines: (a) thick electrode without cover, two discharge rings on $r_1$ and $r_2$; (b) thin electrode without cover, only external ring; (c) thin electrode with a ring-shaped cover, only external ring displaced inside the cover.

Figure 2. Upper view of the discharge in air: (a–c) statical azimuthal inhomogeneities of the discharge ring; (d) transition to the uniform state. Air, $p = 74$ mTorr, $\omega = 9$ MHz, $V_{pp} = 170$ (a), 180 (b), 190 (c) and 200 V (d).

in sputtering systems the external discharge ring is not used and is removed by grounding of electrostatic shield placed close to the electrode. In the present experiment the cylindrical electrostatic shield envelops only the lateral part of the loaded electrode and magnetic system. Therefore, the conditions for sustaining the external discharge are preserved (see figure 1).

For the thin electrode of 2.5 mm in thickness that is in magnetic field with high induction about 0.3 T near $r_1$ the internal discharge ring is not initiated, see figure 1(b). In the third case the dielectric ring was mounted in the place where the external discharge ring is to be initiated, see figure 1(c). At the power less than 5 W the discharge looks as a diffusion ring separated from the teflon cover inside at the distance about 1 mm. Enhancement of the discharge power leads to a glow change: at 10 W the glow ring is pressed to the cover, figure 2(d). At that ring the width decreases in 3 times, the glow intensity inflates which evidently indicates the current density increase. In most part of the investigated gas pressure range at all rf frequencies the transition between glow modes is sharp at some power-level overshoot. It is significant that initiating of dc magnetron discharge in the described conditions turned out to be impossible.

The existence of the third form of the discharge at the power intermediate level 5–10 W is possible, in this form the diffusion ring disintegrates for several segments, figures 2(a–c) and 3(a). When there are more than 6 segments, they are situated with the equidistant spread, as
Figure 3. Rotating azimuthal inhomogeneities of the discharge in air (a) and diffusive glowing ring in argon (b); (a) air, $p = 32$ mTorr, $\omega = 9.2$ MHz, $V_{pp} = 280$ V, $V_{sb} = -80$ V; (b) argon, $p = 37$ mTorr, $\omega = 9.2$ MHz, $V_{pp} = 200$ V, $V_{sb} = -80$ V.

Table 1. Conditions for emergence of azimuth irregularities of the rf discharge in the magnetic field at the power range 5–10 W.

| $\omega$ (MHz) | 5.4 | 6.4 | 9.2 | 13.56 |
|----------------|-----|-----|-----|-------|
| Argon          | 60–80 | 60–80 | --- | --- |
| Argon + 1% air | 30–80 | 30–80 | 30–70 | --- |
| Air            | 30–80 | 30–80 | 20–75 | 34–36 |

Table 2. The electron kinetic coefficients in the rf discharge at $B = 0.1$ T ($\omega_{e,H} = 1.8 \times 10^{10}$ s$^{-1}$), the reduced electric field strength $E/N = 6000$ Td and pressure $p = 80$ mTorr: $N$ is the number density of gas; $\mu_{e,\parallel}$ is the electron mobility along the electric field $E$ and $\mu_{e,\perp}N$ along the vector $E \times B$; $D_e$ is the electron diffusion coefficient; $\nu_{mom}$ is the transport frequency; $k_{ion}$ is the ionization constant; $k_{at}$ is the total attachment constant; $\alpha_{at}$ is the attachment frequency.

| Parameter | Argon | 99% argon + 1% air | Air |
|-----------|-------|--------------------|-----|
| Mean electron energy (eV) | 5.870 | 5.815 | 4.010 |
| $\mu_{e,\parallel}N$ (cm$^{-1}$ V$^{-1}$ s$^{-1}$) | $9.170 \times 10^{18}$ | $9.095 \times 10^{18}$ | $7.628 \times 10^{18}$ |
| $\mu_{e,\perp}N$ (cm$^{-1}$ V$^{-1}$ s$^{-1}$) | $2.406 \times 10^{22}$ | $2.412 \times 10^{22}$ | $1.338 \times 10^{22}$ |
| $D_e$ (cm$^{-1}$ s$^{-1}$) | $8.430 \times 10^{22}$ | $8.133 \times 10^{22}$ | $3.303 \times 10^{22}$ |
| $\nu_{mom}/N$ (cm$^3$/s) | $1.879 \times 10^{-7}$ | $1.863 \times 10^{-7}$ | $1.562 \times 10^{-7}$ |
| $k_{ion}$ (cm$^3$/s) | $7.550 \times 10^{-10}$ | $7.366 \times 10^{-10}$ | $7.643 \times 10^{-11}$ |
| $k_{at} = \alpha_{at}/N$ (cm$^3$/s) | 0 | $6.459 \times 10^{-14}$ | $6.024 \times 10^{-12}$ |

shown in figure 3. The segmented discharge mode is observed at the explicit pressure range $\Delta p$, which depends on the frequency $\omega$. This pressure range becomes wider with the frequency of discharge voltage decreasing and enhancement of air content in the working gas (table 1).

Discharge structures with several evenly spaced segments may rest or rotate around the axis of the symmetry of the system. The stationary rotation was sustained up to tens of minutes. The rotation in the electron $E \times B$ drift direction was observed more frequently in comparison with the inverse one. The conditions that favour to a certain direction are not clear now. The velocity of the inhomogeneities moving along the ring was not more than 10 cm/s.
In table 2, the electron kinetic coefficients in the rf discharge in the magnetic field $B = 0.1$ T for three gases at pressure $p = 80$ mTorr are presented. The electron transport coefficients and rate constants are calculated by BOLSIG+ solver of the Boltzmann equation [14]. We see that the transport coefficients and frequencies and ionization constants weakly vary with gas changing and the attachment constant is negligible in the argon with a small admixture of air. The numerical simulation of the ion composition of the rf discharge plasma in air by the kinetic model [15] shows that at the experimental pressure conditions the main charged plasma species are electrons and $O^+_2$ ions, and the number density of $O^-$ negative ions is very low. So a mechanism of the structure generation in the magnetized rf discharge plasma most likely is not associated with negative ions.

4. Conclusion
In this paper, the axially symmetrical rf magnetron discharge in argon with a small admixture of air and in air was studied. In experiments, up to 10 long-living plasma bunches were found out. They equidistantly located along the discharge ring and could either rotate around the discharge axis or remain motionless. It was concluded that a mechanism of the structure generation in the magnetized rf discharge plasma most likely was not associated with negative ions.

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