Formation of low-frequency periodic structures in a pulsed magnetron discharge

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Abstract. Periodic plasma structures are observed in non-sputtering magnetron discharge (NSMD) that is the transient quasi-stationary low-voltage regime between the high-current magnetron discharge (HCIMD) and an arc. The fast camera imaging synchronized with the magnetic probe diagnostics reveals the correlation between the observed rotation of the plasma inhomogeneities and the magnetic field perturbation behaviour. The frequencies of the periodic processes fall into kHz-range. A simple analytical model of the ionization instability in crossed electric and magnetic fields is suggested for the low-pressure discharge case. Using the model, the possible ranges of wavelengths and frequencies for the plasma inhomogeneities are evaluated. The results show good agreement between the experimental data and theory.

1. Introduction
Recently, several research groups have reported observation and investigation of various wave-like phenomena in high-power impulse magnetron sputtering (HiPIMS) discharges (e.g. [1–3]). Considerable effort is being made to determine the mechanisms of drifting ionization zones appearing in such plasmas, and to gain control over their formation. Our present contribution deals with the investigations of similar effects taking place in quasi-stationary magnetron discharges: high-current impulse magnetron discharge (HCIMD), and low-pressure high-current impulse diffuse discharge (HCIDD), also known as non-sputtering magnetron discharge (NSMD) [4, 5]. HCIMD is a high-voltage (up to 1.5 kV), high-current (up to 20 A/cm²), long (~ 1–40 ms) impulse sputtering tool [4]. The low-voltage (~ 80 V) NSMD dramatically inhibits the sputtering rate thus limiting the deposition process under certain conditions.

Here we consider the experimentally observed periodic structures in case of high-current impulse magnetron discharge on an aluminum target in Ar/CO₂ working gas mixture, and its transition into low-voltage non-sputtering magnetron discharge.

2. Experimental setup
The experiments were carried out in the conventional planar magnetron device as well as in the so-called apparatus with profiled electrodes. The scheme of the experimental setup and the diagnostic instruments is shown in figure 1.
The planar magnetron consists of an 80-mm-diameter disk cathode, grounded anode and the magnetic system. The device with profiled electrodes consists of two hollow axisymmetric Al electrodes arranged between two electromagnetic coils. Current in the coils flows in the opposite directions so that the magnetic field lines form a cusp. The profile of each electrode replicates the magnetic field line curvature, ensuring $E \cdot B \approx 0$ along the most part of their surface. Further apparatus details could be found elsewhere [4, 5].

The discharge power supply system comprises two units: stationary power supply and pulsed power supply. High voltage pulses are generated by commutation of a transmission line using a high-power switch. The energy stored in the transmission line is determined by its charging voltage (e.g. 2 kV corresponds to 3.8 kJ energy). A stationary 0.1 A magnetron discharge provides low density initial plasma.

In the experiments discussed in present contribution the working gas was a mixture of Ar and CO$_2$ with partial pressures $p_{Ar} = p_{CO_2} = 0.5$ Pa.

A fast nine-frame gated camera BIFO K011 is used to perform high-speed imaging of the processes running in the discharge chamber. This device is capable of taking nine images with programmable frame times (100 ns–100 $\mu$s) and frame delays (100 ns–100 $\mu$s) after triggering. The spatial resolution of the matrix is 340×340 pixel for each frame. The spectral sensitivity range is 400–800 nm. Camera was equipped with Zenitar-M 50 mm lens. Simultaneously the Avantes AvaSpec-2048x14 spectrometer was used to record the optical emission spectrum of the plasma thus monitoring the non-sputtering regimes. The electrical parameters of the discharge (current $I_d$ and voltage $U_d$) were recorded by the Tektronix TPS 2024 digital storage oscilloscope. The typical traces of transition from HCIMD to NSMD and then to an arc could be found elsewhere [5].

The fast camera imaging was synchronized with the magnetic probe diagnostics. Magnetic probe was used to measure internal magnetic field perturbations in pulsed plasma. It was calibrated using Helmholtz coils beforehand.

### 3. Experimental results

It was found that the NSMD plasma tends to produce clear optical emission inhomogeneities that rotate along the azimuthal direction. The typical series of fast images made in planar magnetron is presented in figure 2.

The NSMD plasma behaviour was found to be the same in both magnetron devices. The typical FFT spectrum of the magnetic probe signal is presented in figure 3 along with the fast images taken in the apparatus with profiled electrodes (side-on view).
Figure 2. The fast images of NSMD plasma in planar magnetron (frame time 1 µs, frame delay 30 µs).

Figure 3. The low-frequency region of FFT spectrum of the magnetic probe signal (left) and the fast images of NSMD in apparatus with profiled electrodes (right) (frame time 1 µs, frame delay 100 µs).

It appears that the peaks in the FFT spectrum of magnetic probe signal correlate with the observed periodic processes in plasma. Namely, the rotation frequency of the bright regions was estimated as 2–10 kHz both from the fast camera imaging and magnetic probe measurements. Whenever the plasma perturbations were not observed, the low-frequency peaks vanished from the magnetic probe FFT spectra.

4. Model of the sheath processes

Dynamics of the comparable low-frequency processes occurring in Hall thrusters as well as in HiPIMS discharges has been well described in terms of predator-prey models [6, 7]. These models, however, do not consider the nature of the spatial and temporal scales of the azimuthal inhomogeneities. Hence, in order to get the satisfactory description of these phenomena in our experimental conditions we suggest a simple analytical model.

Because the observed periodic processes appear to feature the low frequencies ($f \sim 2$–10 kHz), the proposed model is based on the classical considerations describing the formation of zones with enhanced ionization rate (striations) in the glow discharges [8].

We extend the existing model of the ionization instability [8] to the case of low-pressure plasma in crossed electric and magnetic fields. It is well known that the ionization instability may develop provided the ionization rate depends on the plasma density. Therefore this process is determined by the electron kinetics. Under the conditions typical for magnetron discharges, the cathode sheath is thin and non-collisional, while the inelastic processes that are essential for the discharge existence take place in $\mathbf{E} \times \mathbf{B}$ (azimuthal) direction. Thus in such plasmas the drift-diffusion approach is valid for the azimuthal direction only, and the particles' collisions don't play any role in the other directions. Let us replace a cylindrical system (seen in figure 1) with a Cartesian one and consider a near-cathode region where $\gamma$-axis represents coordinate along the azimuthal direction in a magnetron device. Moreover, let the transverse magnetic field $\mathbf{B}$ be uniform so that electric and magnetic fields are orthogonal in the
whole region considered. Note, that hence the following model is valid for the racetrack zone only. The model of the processes that take place in this simplified near-cathode region is shown in figure 4.

Figure 4. Main elementary processes in the near-cathode region of NSMD.

Here the electrons are emitted from the cathode surface under ion bombardment. Some of them cross the sheath and presheath regions without collisions and may return to the cathode. Those electrons that ionize atoms and molecules in the presheath generate slow secondary electrons that drift in crossed \( E \times B \) field.

Like in a glow discharge case, here the electron drift current is considered as an external one with null divergence. The equation of charged particles balance in plasma appears to be:

\[
\frac{\partial n}{\partial t} + D_{\perp}^{(b)} \frac{\partial^2 n}{\partial y^2} = n v_i,
\]

where \( D_{\perp}^{(b)} = (b_{e,i} D_{e,i} + b_{e,e} D_{e,e})/(b_{e,e} + b_{i,i}) \) is the ambipolar diffusion coefficient across the magnetic field lines [8] (\( b_{e,i} \)— electron and ion mobilities, \( D_{e,i} \)— their diffusion coefficients).

Let us suppose that there’s a small density perturbation in plasma that is periodic along \( y \) axis. Assuming the simple harmonic character of this perturbation the plasma density becomes [8]:

\[
n(x,y,z,t) = n^{(0)}(x,z) + n^{(1)}(x,y,z,t) = n^{(0)}(x,z) + \Re \left[ n^{(1)}_e(x,z,t) \exp(-i k y) \right],
\]

where \( n^{(0)} \) is the initial (unperturbed) plasma density, \( n^{(1)} \) is the small perturbation, \( k \) determines the spatial scale or the wavelength of the perturbation: \( \lambda_k = 2\pi/k \).

The instability develops if the growth rate \( \gamma > 0 \), i.e.

\[
\gamma = n \frac{\partial V_i}{\partial n} - k^2 D_{\perp}^{(b)} > 0.
\]

Taking into account that \( (n/V_i) \partial V_i/\partial n \sim 1 \), we find that if the frequency of ionization \( V_i \) is larger than that associated to ambipolar diffusion (\( k^2 D_{\perp}^{(b)} \)) the plasma density will grow. \( D_{\perp}^{(b)} \) and \( V_i \) may be evaluated using the experimental data. Then the minimal spatial scale of the density perturbation in our model is limited by

\[
\lambda_{k_{\min}} = 2\pi \sqrt{\frac{D_{\perp}^{(b)}}{V_i}}, \quad \lambda_{k_{\min}} = 0.5–5 \text{ cm}
\]

depending on the plasma parameters. The maximal \( \lambda_k \) value can be found after introducing the electron temperature perturbation. Considering the heating of electrons and their energy balance the relation between the electric field and the electron temperature \( T_e \) can be found [8]. Then taking into account the simple harmonic electron temperature perturbation and its relation to the density fluctuation, the balance of charged particles (1) turns into [8]:
\[ \frac{\partial n_e^{(1)}}{\partial t} = \frac{\sqrt{6}}{5} \frac{T_e}{k \lambda_e} \frac{\partial V_i}{\partial T_e} n_e^{(1)} + \left( n \frac{\partial V_i}{\partial n} - D_a^{(0)} k^2 - \frac{6}{5} \frac{T_e}{k^2 \lambda_e^2} \frac{\partial V_i}{\partial T_e} \right) n_e^{(1)}, \]  

where \( \lambda_e \) is the length of electron temperature relaxation, i.e. the path the electron travels until it loses kinetic energy equal to \( T_e \) due to collisions with other particles (we express \( T_e \) in the energy units):

\[ \lambda_e = \frac{b T_e}{e \kappa_{eg} V_{eg}}, \]

where \( \kappa_{eg} \) is the mean fraction of electron energy lost in collisions with neutral gas particles, \( V_{eg} \) — frequency of such collisions [8].

In our case the electrons produce a current along \( y \) axis with current density:

\[ j = en v_a = en \frac{E}{B}. \]

So for the magnetized electrons the modified mobility becomes \( b_e = 1/B \) and hence the temperature relaxation happens on a scale of

\[ \lambda_e = \frac{T_e}{e B \kappa_{eg} V_{eg}}, \]

Solving (6) we derive \( n_i^{(1)} \) and \( n^{(1)} \):

\[ n^{(1)} = \text{Re} \left[ n_i^{(1)} \exp(-i k y) \right] = n \exp(\gamma t) \cos(\omega t - k y), \]

\[ \omega = \frac{\sqrt{6}}{5} \frac{T_e}{k \lambda_e} \frac{\partial V_i}{\partial T_e}, \]

\[ \gamma = n \frac{\partial V_i}{\partial n} - D_a^{(0)} k^2 - \frac{6}{5} \frac{T_e}{k^2 \lambda_e^2} \frac{\partial V_i}{\partial T_e}. \]

The upper limit of the spatial scale of perturbation can be found from

\[ \gamma > 0 \iff k^2 \lambda_e^2 > \left( \frac{T_e}{\frac{\partial V_i}{\partial T_e}} \right) \left( \frac{n \frac{\partial V_i}{\partial n}}{\frac{1}{V_i} \frac{\partial n}{\partial T_e}} \right)^{-1}. \]

Eventually the maximal wavelength is

\[ \lambda_e^{\max} = 2\pi \lambda_e, \quad \lambda_e^{\max} \sim 100 \text{ cm}. \]

Thus the maximal perturbation wavelength is limited by the length of electron temperature relaxation. The latter strongly depends on the prevailing electron energy loss mechanism. Generally speaking, \( \kappa_{eg} V_{eg} = \kappa_{eg}^{(el)} V_{eg}^{(el)} + \kappa_{eg}^{(inel)} V_{eg}^{(inel)} \), where superscripts (el) and (inel) correspond to elastic and inelastic processes. So the electrons with energies lower than the inelastic processes threshold mostly lose their energy in elastic collisions with atoms and molecules. On the contrary, electrons with rather high energies sufficient for excitation and/or ionization processes predominantly lose their energy in inelastic collisions. Nevertheless their frequency is lower, the fraction of energy lost is much higher for inelastic processes.

Then the situation may be clarified if we evaluate the energy electrons gain on their cycloidal paths: \( E_c = e E, h = 2m_e E^2_c / B^2 \). The internal electric field in the presheath is experimentally measured: \( E_c \sim 10^4 \text{ V/m} \). The magnetic field decreases from 0.03 T to 0.01 T when moving away from the cathode surface across the sheath and presheath.
Then the electrons drifting close to the cathode surface (in the lower region in figure 2) gain $\varepsilon_{\text{ec}} \approx 1\text{eV}$. In this case the elastic losses dominate, hence $\nu_{\text{eg}} \approx \nu_{\text{eg}}^{(\text{el})}$ and $\lambda_{\text{max}}^{(\text{el})} = 2\pi \lambda_{\text{e}} \approx 100\text{ cm}$.

On the contrary, electrons drifting in the presheath (in the upper region in figure 2) gain $\varepsilon_{\text{ec}} \approx 10\text{eV}$. Here $\nu_{\text{eg}} \approx \nu_{\text{eg}}^{(\text{inel})}$ and the maximal wavelength decreases: $\lambda_{\text{max}}^{(\text{inel})} = 2\pi \lambda_{\text{e}} \approx 10\text{ cm}$.

Our evaluations of the energies are valid for the average values only. Obviously, both the elastic and inelastic processes take place in the whole presheath while the accurate $\nu_{\text{eg}}^{(\text{inel})}$ value must be calculated using the electron energy distribution function. Nevertheless, our approach allows determining upper and lower limits for the ionization instability wavelength.

The spatial parameters of the experimentally observed plasma inhomogeneities match those evaluated using the simple model, i.e. $1\text{ cm} < \lambda << 100\text{ cm}$. The frequency of the perturbation wave according to (9) gives $f = \omega/(2\pi) = 10^2$–$10^4\text{ Hz}$. Thus both the spatial and time scales of the observed wave phenomena in NSMD are in good agreement with the theoretical results. The proposed analytical model may be also adopted for describing the plasma dynamics in high-power sputtering magnetron modes (HCIMD and HiPIMS).

5. Conclusion
The low-frequency (2–10 kHz) periodic structures are observed and investigated in non-sputtering magnetron discharge operated on Al target in oxygen-containing working gas mixture. A simple analytical model of the ionization instability in crossed electric and magnetic fields is proposed. Experimental results are in good agreement with theory.

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