Dusty waves and vortices in rf magnetron discharge plasma

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Abstract. The appearance and subsequent growth of metallic particles in plasma of planar rf magnetron sputter were observed. The origin of the particles is sputtering of the rf electrode by ion flux from the plasma. In some regions of formed dust cloud the particles were involved in the horizontal or vertical circular movement. The horizontal rotation along the sputtered track in the cyclotron drift direction was observed close to the main magnetron plasma. The torus-shaped dust vortex ring engirdled the secondary plasma of the discharge at height of a few centimeters over the electrode. Close to this region particle density waves propagated through the cloud. The possible role of discharge plasma azimuthal inhomogeneity and gas dynamics effects in the forming the observed structures was considered.

1. Introduction
The investigations of dust waves in dusty plasma began in the early nineties with the works, where excitation of dust acoustic waves was analyzed [1, 2]. In [3] the dispersion relation for the dust wave was obtained basing on hydrodynamic model of the dusty plasma. The theoretical and experimental investigations were actively carried on later, the results may be found in the book [4]. In [5] the different types of instability were analyzed which develop in the simultaneous presence of both gradient of particles charges in the cloud and of arbitrary nonelectric field. Authors distinguished the type that leads to dust vortex appearance whether or no the dampening factor is essential. The latest investigations of the dust instabilities are represented in [6–8].

Relating the topic of the present paper it is necessary to note the work [9] where the low-frequency fluctuations were discovered in the dust cloud that grew in the reactive plasma of hollow post magnetron system. The particle density fluctuations developed simultaneously with the electron density ones, were antiphase to them and appeared only on reaching rather high density of the micron-sized particles in plasma.

Magnetron sputtering at elevated pressure of 10–100 Pa is used to obtain the metallic clusters with diameters of some nanometers [10]. The techniques for obtaining larger particles by
Figure 1. The schematic side view radial cross-section of the electrode system and the particle clouds in the plasma: 1—the sputtered target (the small arrows symbolize sputtered atom flux); 2—the substrate; 3—the earthed shield; 4—the magnetic system; 5—the discharge glowing ring; 6 and 7—the particle clouds.

sputtering in the rf plasma with magnetic field and without it are presented in [11] and [12] correspondingly.

The formation of the trap for the particles in the rf magnetron plasma over the flat electrode was realized in [13]. According to experimental results of [13] the cloud of the particles may be trapped in the plasma nearby the sputtered track during the sputtering process. Trapping of the dispersed particles in plasma close to the sputtered target may be used for coating them by metal and farther synthesis of composite materials [13–16].

The volumetric dust cloud consisting of particles that grew and were confined in the magnetron plasma over the sputtered electrode was observed in [17]. The aim of the present work was the investigation of spatial-temporal processes in such cloud.

2. Experiment

The axisymmetric planar magnetron sputter equipped with the flat copper water cooled target 1 (figure 1) and with the unbalanced magnetic system 4 was positioned in the middle of vacuum chamber.

The maximal radial magnetic induction value was 0.03 T. Plasma over the target was maintained by capacitive coupling to 5.28 MHz rf supply. The target served as the live rf discharge electrode and the chamber walls did as the earthed one. The rf peak-to-peak voltage was 400–480 V, argon gas pressure $p$ was 6.5–50 Pa, gas flow rate was 5 sccm. The live electrode self-bias was close to zero.

The clouds of the particles that nucleated and grew in plasma were visualized by brightening with the 300 mW laser beam. By means of cylindrical lens the beam was expanded to sheet that was positioned vertically or horizontally. The images were recorded by photo camera and video camera at 200 fps. The processes were interrupted in different studies of particle growth in the range of 15–660 min from the beginning. During the discharge switching-off the grown particles were being collected onto the special substrates. Before the switching-off the positive potential of 135 V was supplied to the substrate 2 (figure 1). The sedimentation process was visualized and recorded too. Another substrate was positioned on the bottom of the chamber. The SEM (scanning electron microscope) images of the particles were obtained and their elemental composition was investigated by EDX (energy-dispersive X-ray spectroscopy). The average concentration of particles $n_d$ in the cloud was estimated by measuring the laser light extinction taking into account the particle size obtained from the SEM images. The emission spectra of ring-shaped main plasma glow 5 (figure 1) were obtained with a scanning
monochromator. Spectral composition of rf voltage was investigated with built-in FFT (fast Fourier transform) function of TDS 2024 oscilloscope.

3. Results and discussion
At the initial stage of sputtering, namely during the first 30 min, the discharge glow was keeping the stable contours, its intensity was changeless in time and uniform azimuthally. The side view of the magnetron discharge glow is given in figure 2.

In contrary the various types of periodic changes were proper to the plasma glow at the later stages of sputtering. Firstly, the regions of raised glow intensity (or bunches) were observed in the brightest glowing ring region located close to the electrode. In plasma located over the ring the corresponding to them bunches having less brightness were observed. There were from 10 to 20 bunches simultaneously in the ring. They were distributed regularly along the ring and rotated synchronously in the $E \times B$ electron drift direction with the frequency of about 3 Hz. In such a way in any cross-section of the plasma ring the glow intensity vibrated at frequency in the range of 30–60 Hz.

Beside this the slow radial pulsations of the plasma contours were observed after some hours from the sputtering beginning. It look as the serial enlargement and shrinkage of the glowing region volume in the radial direction. The volume enlargement was followed by the glow intensity one.

The observations showed that the described changes of the discharge glow are interconnected with appearance of the large dusty cloud in the vacuum chamber and with its dynamic features. During the sputtering process at the argon gas pressure in the range of 19–50 Pa the appearance and subsequent growth of particles in the definite plasma regions (traps) were observed. The visual observing the particles became possible after 2–7 min of the sputtering process beginning. The analysis of the collected particles showed that the particles consisted of the target material—copper. The particles formed the clouds in the traps. The particles that were collected from different clouds showed different crystalline structures. In the text therein under only the results obtained at $p = 19–21$ Pa are presented. During the first hour of sputtering process the initial cloud looked as a ring coaxial to the discharge glowing one, and located close to or inside the plasma at height of $d = 3–4$ mm over the target (see 6 in figure 1 and figure 3(a)). The particles in the clouds rotated around the target center in the direction of the electron drift in the crossed fields [6].

The size distributions of the particles, that were collected from initial cloud at different points of time, were nearly monodisperse ones. The mean particle size in this stage increases with time almost linearly with the growth rate of 10 nm/min, see figure 4.
Figure 3. Typical view of the discharge plasma glow (lilac, native) and the cross-section of the particle clouds in plasma (visualized with the green laser): at the initial stage of sputtering \((a)\) and at the later one \((b)\). Photo camera.

Figure 4. Time dependence of the mean size of copper particles grown in the plasma.

There were no agglomerates among the submicron particles. Not more than 5% of the particles having the sizes about 1 \(\mu m\) aggregated forming the pairs in plasma (see figure 5, left). The particles of 0.1–1 \(\mu m\) sizes grown in the cloud had crystalline faceting; a lot of particles were single crystals or twins with the shape, typical for fcc lattice (figure 5, right).

As the particles reached 1 \(\mu m\) size, the particle flow from the initial cloud raised toward the discharge periphery. Over the earthed shield electrode the secondary cloud (7 in figure 1) was formed from these particles. In several hours after the discharge was turned on this cloud reached the volume of \(10^3\) cm\(^3\). According to SEM images of the particles collected on the substrate placed on the bottom of the vacuum chamber, the particle size in this secondary cloud remained as much as about 1 \(\mu m\) and did not increase. The cross-section of such cloud is shown in figure 2\((b)\) and figure 7. Average particle concentration in this cloud reached about \(10^6\) cm\(^{-3}\).
On the contrary, in the initial cloud (close to sputtered electrode) the particle growth continued. In the later stage of the growth the particle agglomeration with forming chains of particles up to 500 μm length was observed figure 6.

The majority of the particles in both the clouds were in motion. Particles in the initial cloud rotated in the horizontal plane along the sputtered track in the cyclotron drift direction. Only in the periphery of the cloud the particles were still and formed the plasma crystal.

A part of the particles in the secondary cloud located close to the discharge boundary was involved in the vortex movement. The vortex ring had the form of the torus engirdled the secondary plasma of the discharge at height of about 3 cm. If the quantity of the particles in the cloud is not too high, the particles concentrated within the torus. The left part of such symmetric vortex ring is shown in figure 7. The directions of arrows coincide with ones of instantaneous particles velocities, and their lengths are proportional to the velocity values. The parallel to each other velocities in the bottom of the vortex pattern corresponds to transition to the cloud pattern comprising plane dust waves. The distribution of the velocities was obtained with PIVlab software [18]. It is known that if there are small particles in gas in vicinity of a gas dynamic vortex ring, their maximum concentration is near the azimuthal axis of the ring [19]. One can suppose thereupon that in our plasma the gas vortex really did located in the region of the dust vortex coaxial to the latter. The location of the ring in the larger cloud is pointed in figure 8.

The diameter of the torus changed periodically just in antiphase with the observed slowest vibrations of circular border of discharge glow.

From the internal border of the secondary cloud to the periphery the particle density waves propagated through the cloud. They may be observed in the image of the cloud obtained by laser light scattering as alternation of bright and dark fringes (see figure 8). Simultaneously two wave patterns may be observed which propagated at oblique angles to each other direction. The vibration frequency in the longer wave is approximately of 30 Hz and in the other one is of 60 Hz. Propagation of the longer wave periodically deformed the bottom border of the secondary cloud.

**Figure 5.** Typical view of the collected copper particles grown in plasma for 110 min. SEM.
Figure 6. Typical view of the collected agglomerates of particles grown in plasma for 10 hours. SEM.

The major input into vibrations of the scattering light intensity gave the somewhat divergent waves having larger length. They propagated in the direction close to those one marked in figure 8 with straight line. The luminance of cloud image was measured in the points of the line. The luminance variation along the line is given in figure 9.

The wavelength $\lambda$ measured as the average distance between the crests in figure 9 is of 0.3 cm. The phase velocity $C = \lambda \nu = (0.3 \text{ cm}) \times (30 \text{ Hz}) = 9 \text{ cm/s}$. In the moment we may estimate only roughly the dust wave phase velocity in frames of hydrodynamic model of the plasma. Following the model [20] the velocity $C_{\text{HM}} \approx \omega_{pd} \lambda_{D, \text{ eff}}$, where $\omega_{pd} = [n_d e^2 Z^2/(\varepsilon_0 m_d)]^{1/2}$ is the dust plasma frequency and $\lambda_{D, \text{ eff}}$ is the effective charge screening length that may be taken equal to the ion Debye length; $Ze$ is the charge of particle; $m_d$ is particle mass. As result $C_{\text{HM}} \approx P_{H}[n_i k_B T_i/(m_i n_d)]^{1/2}$, where $P_{H} = Z n_d / n_i = (n_i - n_e) / n_i$ is the Havnesfl parameter, $n_i(e)$ is ion (electron) number density, $T_i$ is the ion temperature. To estimate $Z$ we take in account that the described wave pattern located in plasma periphery region having almost no visual optical emission (see figure 3), so we consider the value in the region of 0.5–1 eV as the upper estimation of $k_B T_e$, where $T_e$ is the electron temperature. Then $Z$ is equal to 1000 or somewhat less, which lead to Havnesfl parameter $P_{H}$ close to unity. Taking the volume density 8.9 g/cm$^3$ (copper) for particle mass calculation one may obtain $C_{\text{HM}} = 3 \text{ cm/s}$. The difference between $C_{\text{HM}}$ and measured $C$ values seems to be related both to inaccuracy of the effective charge screening length and to the overestimation of the particle concentration by means of laser light extinction. The latter may be due to quite large optical path of the laser beam through the cloud. Accurate estimate may be fulfilled basing on the probe measurements of the plasma...
parameters and on the measurements of particle number density by means of the high resolution cloud images, which we plan to carry out.

The observed movement of alternating bright and dark patterns in the plasma ring glow argues for the periodical change of plasma parameters in any fixed point of the plasma ring. Some attributes indicate that namely the plasma bunches are the sources of the dust wave appearance
Figure 9. The luminance variation in image of the cloud along the line marked in figure 8.

in the cloud that surrounded the glowing region. The attributes are as follows: simultaneous development of both the wave propagation process and the plasma bunches movement, the observed features of wave pattern location and the wave propagation direction and coincidence of the wave and frequency ranges. The azimuthal movement of the bunches produces the periodical perturbation of plasma parameters in region of the inner border of dusty cloud and corresponding periodical changing of the particles charge. This changing causes the vibrations of the particles at the border relative to neighboring ones which propagate over the cloud as the dust waves. In turn, one may state that the plasma bunches appearance is initiated by filling the ionization region of the discharge with dust particles of quite large size. The uniformity of the discharge glow at the initial stage of sputtering, when the particles are small, suggests this.

4. Conclusion
In summary, the following was observed in planar rf magnetron sputter discharge plasma under elevated pressure of tens of pascals: the growth of micron-sized dust particles from the sputtered copper atoms; formation of the large dense dusty cloud; confinement of the cloud in plasma for a long time of some hours or more; the pulsations of particles concentration in the cloud propagating through it as the waves having phase velocity about 10 cm/s. Two wave patterns having different wavelengths are clearly distinguishable. The dusty particles presence in the magnetron discharge plasma ionization ring-shaped region produces the stratification of the plasma. The moving plasma bunches are the source of the longer dust wave in the cloud. The large volume of the cloud allows observing other occurrences of the dusty medium dynamics such as the torus shaped vortex ring with the torus generatrix diameter of a few centimeters.

Acknowledgments
This work was supported by the Russian Science Foundation (project No. 16-12-10424).
References

[1] Rao N N, Shukla P K and Yu M Y 1990 *Planet. Space Sci.* 38 543–6
[2] Rosenberg M 1993 *Planet. Space Sci.* 41 229–33
[3] Kaw P K and Sen A 1998 *Phys. Plasmas* 5 3552–9
[4] Shukla P K and Mamun A A 2001 *Introduction to Dusty Plasma Physics* (Berlin: CRC Press)
[5] Vaulina O S, Samarian A A, Petrov O F, James B and Melandso F 2004 *Plasma Phys. Rep.* 30 918–36
[6] Rosenberg M 2016 *IEEE Trans. Plasma Sci.* 44 451–7
[7] Jana S, Banerjee D and Chakrabarti N 2016 *Phys. Lett.* A 380 2531–9
[8] Wang Y L, Guo X Y and Li Q S 2016 *Commun. Theor. Phys.* 65 247–53
[9] Chu J H, Du J B and Li L 1994 *J. Phys. D: Appl. Phys.* 27 296–300
[10] Kashtanov P V, Smirnov B M and Hippler R 2007 *Phys. Usp.* 50 455–88
[11] Terauchi S, Koshizaki N and Umehara H 1995 *Nanostruct. Mater.* 5 71–8
[12] Samsonov D and Goree J 1999 *J. Vac. Sci. Technol., A* 17 2835–40
[13] Rudavets A G, Ryabinkin A N and Serov A O 2011 *Plasma Processes Polym.* 8 346–52
[14] Ekimov E A, Borovikov N F, Ivanov A S, Pal A F, Rusinkevich A A, Ryabinkin A N, Serov A O, Starostin A N, Fortov V E and Gromnitskaya E L 2014 *Diamond Relat. Mater.* 41 1–5
[15] Ekimov E A, Ivanov A S, Pal’ A F, Ryabinkin A N, Serov A O, Starostin A N, Fortov V E, Sadykov R A, Mel’nik N N and Presh A 2005 *Dokl. Phys.* 50 351–4
[16] Ivanov A S, Mitin V S, Pal’ A F, Ryabinkin A N, Serov A O, Skryleva E A, Starostin A N, Fortov V E and Shulga Y M 2004 *Dokl. Phys.* 49 163–6
[17] Filippov A V, Pal A F, Ryabinkin A N and Serov A O 2013 Growth of particles from sputtered metal in an rf magnetron discharge plasma trap XXVIII Int. Conf. Interaction of Intense Energy Fluxes with Matter (Elbrus, Kabardino-Balkaria, Russia) pp 175–8
[18] Thielicke W and Stamhuis E J 2014 *J. Open Res. Softw.* 2 e30
[19] Munro R J, Bethke N and Dalziel S B 2009 *Phys. Fluids* 21 046601
[20] Merlino R L 2014 *J. Plasma Phys.* 80 773–86