Spectroscopic evaluation of the effect of the microparticles on radiofrequency argon plasma

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Abstract. Axial distributions of 1s excited states of argon were measured in a radiofrequency (RF) discharge by a self-absorption method. Experiments were performed in the PK-3+ chamber, designed for microgravity experiments in complex (dusty) plasmas on board the International Space Station. A correction of a standard self-absorption method for the extinction of the light by the levitating microparticles is proposed. Distributions, measured at the same discharge conditions in a microparticle-free discharge and a discharge containing a cloud of levitating microparticles, revealed the non-local influence of the microparticle cloud on the discharge plasma. The most probable cause of this influence is the disturbance of the ionization balance by the levitating microparticles.

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1. Introduction

Complex (dusty) plasmas are of great interest for technological applications as well as from the point of view of basic physics. Many industrial processes suffer from the presence of microparticles in the volume of discharges [1] and in many of them the presence of microparticles is desirable [2]. For fundamental science complex plasmas represent unique models of strongly coupled systems, available for observation at a kinetic level [2, 3]. For both fields, the influence of microparticles on the plasma is of significant importance. It has been known for a long time that the losses of plasma particles on the surface of the microparticles suspended in a plasma, may be comparable to that on the walls of the setup, resulting in e.g. faster decay of the plasmas [4]. Later, in the 1990s the influence of nanometer-sized particles on capacitively coupled radiofrequency (RF) discharges was extensively studied. The main results of these studies can be summarized as follows: the presence of nanometer-sized particles with a density of the order of $10^{14} \text{m}^{-3}$ in a plasma leads to a strong shift of the ionization balance in the discharge. This shift reveals itself in changes of practically all the plasma properties: current–voltage characteristics and RF-matching parameters [5, 6], increase of integral intensity of the radiation as well as the number density of metastables [5, 7], dramatic decrease of the electron density and increase of temperature [5, 6]. This transformation was given the name ‘$\alpha-\gamma$’ [5, 8, 9] transition in analogy with the $\alpha-\gamma$ transition previously known for capacitively coupled RF discharges [10]. Recently, such tiny effects as the modification of the $H_\alpha$ line shape in the presence of nanometer-sized particles have been revealed [11].

For nanometer-sized particles, the gravitational force is negligibly small. Therefore, if injected into or grown in the discharge, these particles will occupy the entire plasma volume. It is not surprising then that their influence on the discharge has a global character. A very local effect of microparticles on the plasma has been recently demonstrated by Do et al. [12]. They levitated a very thin (2–3 mm vertical extension) cloud of monodisperse silica microspheres of 10 $\mu\text{m}$ diameter and measured the axial and radial profiles of the number density of metastable...
states using laser absorption spectroscopy. The microparticle-induced changes were very local. The density of metastables decreased (in contrast to the increase in the case of experiments with nanometer-sized particles) only inside the microparticle cloud. This decrease was attributed to the loss of metastables on the surface of the suspended microparticles.

In this work, we concentrate on the evaluation of the effect of microparticles on the plasma in the PK-3+ chamber, which is the heart of the International Space Station-based complex plasma laboratory [13]. In several microgravity and ground-based experiments in PK-3+ [13] and its predecessor PK-3 Nefedov [14, 15], changes of the plasma glow distribution, caused by the motion of microparticles (e.g. injection of microparticles, heartbeat instability, etc), were observed. This suggests the necessity for the evaluation of the effect of microparticles on the plasma in this device. We use a single-mirror self-absorption method [16, 17] to measure the absolute values of the number densities of metastable and resonant states of argon in the presence of microparticles as well as in the absence of microparticles in the discharge. We try to draw conclusions about the physical mechanisms underlying the observed microparticle-induced changes.

2. Experimental method

2.1. Experimental setup

The PK-3+ setup is, briefly, a symmetrically driven parallel plate 13.56 MHz RF discharge with disc-shaped electrodes 6 cm in diameter and 3 cm gap. Experiments were carried out at working pressures of 15, 30 and 60 Pa with argon buffer gas. RF voltage of about 20 V peak-to-peak was applied to the electrodes. RF power of 0.2 W was kept constant in all the experiments. Electron temperature and density lie in the range of 3–7 eV and $10^{14} - 10^{15} \text{ m}^{-3}$ [18, 19].

Light from the discharge was collected by an optical fiber with a collimator attached to it. The collimator provided the horizontal line of sight of approximately 2 mm thickness. A Hamamatsu Mini-spectrometer TM-series with 3 nm resolution was used to obtain the spectrum. The optical fiber could be moved vertically by a translation stage in order to obtain the vertical profile of light emission (figure 1). Spectra were recorded with a 3 mm step.

For the measurements of self-absorption a protected-Al mirror, opposing the fiber, was used. For each position of the fiber and for each discharge condition, spectra with and without the mirror were recorded. Also the measurements were repeated twice at the same argon pressure for the microparticle-free plasma and the plasma with a large cloud of microparticles, levitating in it.

As microparticles we used monodisperse plastic (melamineformaldehyde) microspheres of 2.55 μm diameter. To observe them levitating in a plasma we illuminated them with a vertical laser sheet and observed the scattered light with a video camera (figure 1). The number density of the microparticles was always of the order of $10^{11} \text{ m}^{-3}$. The same videocamera could be used for imaging of the integral glow of the discharge.

In the PK-3+ setup, the refreshment of gas is performed in pulses. During all measurements the time interval between two pulses was set to 40 s. This periodical gas refreshment resulted in a slight periodic change of discharge parameters. This fact was reflected in the periodical behavior of the intensities of the spectral lines. Therefore, the spectral measurements were synchronized with the gas injection and were performed 20 s after it.

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Figure 1. Scheme of the experimental setup. A discharge is generated by applying RF voltage to the electrodes. Under the laboratory conditions microparticles levitate closer to the bottom electrode due to gravity. They are illuminated with a vertical laser sheet and observed by a video camera. An optic fiber is used to collect the light from the discharge. A mirror, placed opposite to the fiber, allows the evaluation of self-absorption.

2.2. Determination of the number density of the argon states

The populations of the argon levels are determined using a well-known single mirror self-absorption method [16, 17]. Discharge radiation, reflected by the mirror, is used to probe the plasma. Measurements of the intensities of the spectral lines for the transition \( i \rightarrow j \) with and without the mirror allow us to determine the relative absorption \( A_{ij}^L \):

\[
A_{ij}^L = \frac{1 + r_{ij} - I_{ij}^m / I_{ij}^n}{r_{ij}},
\]  

(1)

where \( r_{ij} \) is the reflectance of the mirror at the wavelength \( \lambda_{ij} \) of the considered transition, \( I_{ij}^m \) and \( I_{ij}^n \) are the line intensities with and without the mirror, respectively. We assume that our plasma is uniform along the (radially directed) line of sight, i.e. the top and bottom states of each radiative transition as well as the microparticles are distributed uniformly. At the same time, we consider axial distributions of the parameters to be important. In this approximation, the intensity of a spectral line registered by a detector, observing a certain line of sight, may be expressed as follows [16]:

\[
I_{ij} = \int_{-\infty}^{\infty} \frac{I_{ij}^L(v)}{\kappa_{ij}(v)} (1 - e^{-\tau_{ij}(v)}) dv,
\]  

(2)

where \( v \) is the frequency detuning from the center of a spectral line, \( I_{ij}^L(v) \) is the intensity profile, emitted by the unit of length of the plasma, \( \kappa_{ij}(v) \) is the absorption profile, so that the optical thickness of the plasma \( \tau_{ij}(v) = \kappa_{ij}(v) \times l \) (\( l \) is the length of the plasma along the line of sight). The mirror effectively serves as a light source with the intensity \( I_{ij}^L = r_{ij} I_{ij} \), of which the intensity

\[
I_{ij}^L = r_{ij} \int_{-\infty}^{\infty} \frac{I_{ij}^L(v)}{\kappa_{ij}(v)} e^{-\tau_{ij}(v)} \left(1 - e^{-\tau_{ij}(v)}\right) dv
\]  

(3)

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will be transmitted through the plasma to the detector side. The relative absorption is then $A_{ij}^L = (I_{ij}^L - I_{ij}^s)/I_{ij}^s$. Substituting equations (2) and (3) into the expression for $A_{ij}^L$ after some algebra yields

$$A_{ij}^L = 2 - \frac{\int_{-\infty}^{\infty} \frac{I_{ij}^L(v)}{\kappa_{ij}(v)} \left(1 - e^{-2\tau_{ij}(v)}\right) dv}{\int_{-\infty}^{\infty} \frac{I_{ij}^L(v)}{\kappa_{ij}(v)} \left(1 - e^{-\tau_{ij}(v)}\right) dv}. \quad (4)$$

In microparticle-free plasmas, the only mechanism contributing to $A_{ij}^L$ is the absorption of the radiation by the bottom states of respective transitions, i.e. self-absorption. In addition to that, in complex plasmas microparticles will also effectively contribute to the absorption due to extinction. Therefore total absorption coefficient and optical thickness may be rewritten as a sum of plasma and microparticle components: $\kappa_{ij}(v) = \kappa_{ij}^d(v) + \kappa_{ij}^{pl}(v)$ and $\tau_{ij}(v) = \tau_{ij}^d + \tau_{ij}^{pl}(v)$. The microparticle contribution may be considered to be independent of the frequency within a typical width of a spectral line since the variation of the Mie-scattering and absorption cross-sections as well as the refractive index of the microparticle material is negligible in such a narrow spectral band. Equation (4) may therefore be rewritten in the following way:

$$A_{ij}^L = 2 - \frac{\int_{-\infty}^{\infty} \frac{I_{ij}^L(v)}{\tau_{ij}^{pl}(v) + \ln(K_{ij}^d)} \left(1 - e^{-2\tau_{ij}^{pl}(v)}\right) dv}{\int_{-\infty}^{\infty} \frac{I_{ij}^L(v)}{\tau_{ij}^{pl}(v) + \ln(K_{ij}^d)} \left(1 - e^{-\tau_{ij}^{pl}(v)}\right) dv}. \quad (5)$$

Here $1/K_{ij}^d = \exp(-\tau_{ij}^d)$, i.e. $K_{ij}^d$ is the attenuation of a spectral line by the microparticles levitating in a plasma.

Profiles $\kappa_{ij}^{pl}(v)$ and $I_{ij}^L(v)$ are determined by the broadening mechanism of a spectral line. Estimations, using the experimental data on the broadening of Ar lines [20, 21], show that under our conditions collisional broadening is negligible and consequently a pure Doppler profile can be used: $\kappa_{ij}^{pl}(v), \tau_{ij}^{pl}(v), I_{ij}^L(v) \propto \exp(-\lambda_{ij}^2 M^2 / 2k_BT v^2)$, where $M$ is the argon atom mass and $T$ is the temperature of the neutral gas. This allows us to bind the relative absorption with the optical thickness in the center of the line $\tau_{ij}^{pl}(0)$:

$$A_{ij}^L = 2 - \frac{\int_{-\infty}^{\infty} \frac{e^{-\omega^2}}{\tau_{ij}^{pl}(0)e^{-\omega^2} + \ln(K_{ij}^d)} \left(1 - \frac{\exp(-2\tau_{ij}^{pl}(0)e^{-\omega^2})}{(K_{ij}^d)^2}\right) d\omega}{\int_{-\infty}^{\infty} \frac{e^{-\omega^2}}{\tau_{ij}^{pl}(0)e^{-\omega^2} + \ln(K_{ij}^d)} \left(1 - \frac{\exp(-\tau_{ij}^{pl}(0)e^{-\omega^2})}{K_{ij}^d}\right) d\omega}. \quad (6)$$

where $\omega = \lambda_{ij} \sqrt{M / 2k_BT} v$ is a dimensionless integration variable. In the absence of microparticles ($K_{ij}^d = 1$) expression (6) is reduced to a well-known equation for a single mirror method [17].

On the other hand, if self-absorption is switched to zero ($\tau_{ij}^{pl}(0) = 0$), equation (6) reduces to $A_{ij}^L = 1 - (K_{ij}^d)^{-1}$, which represents the relative absorption of a monochromatic wave [16]. The latter is expected, since we neglected the detuning dependence of $K_{ij}^d$. The stronger the
Figure 2. Dependence of the relative absorption $A_{ij}^L$ on the optical thickness in the center of a spectral line $\tau_{ij}^{pl}(0)$ at different attenuations by microparticles $K_{ij}^{mp}$. The higher $K_{ij}^{mp}$ is, the smaller $\tau_{ij}^{pl}(0)$ is required to provide the same total relative absorption. The contribution of microparticles to the total absorption is especially significant at small $\tau_{ij}^{pl}(0)$.

All the quantities in the rhs of (1) can be experimentally measured. Therefore $\tau_{ij}^{pl}(0)$ can be determined from (6), provided the respective microparticle-caused attenuation $K_{ij}^{d}$ is known.

Supposing the Doppler broadening of the line, $\tau_{ij}^{pl}(0)$ is connected with the number density of the bottom state of the transition [22]:

$$\tau_{ij}^{pl}(0) = \frac{\lambda_{ij}^3}{8\pi} \sqrt{\frac{M}{2k_B T}} \frac{g_i}{g_j} A_{ij} n_j,$$

where $g_i$ and $g_j$ are the statistical weights of the top and bottom states of the transition, respectively, $A_{ij}$ is the Einstein coefficient and $n_j$ is the number density of a bottom state.

Using the experimental method described above and solving equations (1), (6) and (7) for the following spectral lines of argon: 706.7, 794.8, 738.3 nm and 826.4 nm, we could determine the populations of the 1s$_5$, 1s$_3$, 1s$_4$ and 1s$_2$ levels, respectively.

2.3. Estimation of the microparticle-caused attenuation

To correctly determine the number density of argon states in complex plasmas, we need to take into account the extinction of the plasma radiation by the microparticles levitating in it. It is, however, difficult to estimate the extinction cross-section numerically due to the lack of data on the refractive index of melamineformaldehyde in the spectral range of interest. Clean in situ measurement is also a problem due to the difficulty in distinguishing between the extinction and self-absorption.

We estimated the extinction experimentally. To perform this, we ignited a neon discharge in the same PK-3+ chamber and levitated in it clouds of the same microparticles, with the number...
Figure 3. Measured spectral dependence of the attenuation $K_{ij}^d$ and relative absorption $1 - (K_{ij}^d)^{-1}$ of light by a cloud of microparticles. In the spectral range of interest, the relative absorption varies by more than a factor of two. The highest value of $K_{ij}^d$ obtained in our experiments is $\approx 1.13$.

densities and axial extensions close to those that we observed in the main experiment. We used an argon lamp to evaluate the attenuation of its light by the cloud of microparticles. In this case, self-absorption does not take place. Therefore we may assign $K_{ij}^d$ the attenuations measured in this way. Typical values of $K_{ij}^d$ for our dust clouds (microparticle density of $\sim 10^{11} \text{ m}^{-3}$) are given in figure 3.

3. Results

3.1. Microparticle-free plasma

Here we present the results of the measurements in the microparticle-free plasma. Since the discharge is strongly non-uniform along its axis $z$, for the correct comparison of different regimes we always consider axial distributions of the quantities of interest. The point $z = 0$ corresponds to the bottom electrode. In all the experiments, measured line intensities followed the classical glow distribution in the so-called $\alpha$-form of a RF discharge [10]: they had humps close to the powered electrodes and a dip in the center. Examples of these distributions for the argon pressure of 15 Pa are shown in figure 4. Optical thicknesses and number densities exhibit similar behavior. Maximal values of optical thicknesses are of the order of unity and are therefore not negligible, allowing for the determination of the line-of-sight integrated number densities $n_j \times l$. Remarkable is the hierarchy of number densities of different levels, also independent of pressure: the $1s_3$ level has the smallest number density, significantly larger and very close to each other are those of two radiative levels and $1s_5$ exhibits the largest population. The same hierarchy was obtained in a GEC reference cell by the so-called ‘robust method’ [22] at argon pressures higher than 1 Pa.

Axial distributions of $n_j$ are pressure dependent. All the levels behave similarly with pressure. An example for a metastable level $1s_5$ and radiative level $1s_2$ is given in figure 5. In the humps, the number densities are considerably larger for 60 Pa than for 15 and 30 Pa. In the central area, the tendency is opposite: at 15 Pa the number densities are increased with respect to those at 30 and 60 Pa. The pressure dependence of the number densities is summarized in
Figure 4. Axial distributions of intensities and optical thicknesses for the considered spectral lines and line-of-sight-integrated densities of the respective bottom levels at an argon pressure of 15 Pa in a microparticle-free discharge. All three quantities exhibit similar behavior with humps close to the electrodes and a dip in the center.

table 1: absolute maximal values and minimal values in between the humps at different pressures are given.

3.2. Effect of a microparticle cloud

The effect of the levitating cloud of microparticles on the plasma can be seen from the simple imaging of the discharge glow. Figure 6(a) represents the image of a microparticle-free discharge. The glow in this case is symmetric with respect to the middle plane of the discharge. Injection of microparticles leads to the evident break of this symmetry. Due to gravity microparticles concentrate themselves in the glow region above the bottom electrode (figure 7(a)) and as a result the shape of this glow region is significantly modified—it is broadened and extends significantly deeper into the central, relatively dark area of the discharge.
Figure 5. Pressure dependence of the axial profiles of the number densities for 1s₂ and 1s₅ states of argon. Humps of the distributions grow on the increase of pressure, whereas the dip, on the contrary, considerably drops as the pressure is increased. Similar dependence on pressure is observed for all four considered levels.

Table 1. Line-of-sight integrated number densities of metastable and radiative states of argon in the PK-3+ chamber at humps and dips of the axial distribution for different pressures. The values are given in $10^{15}$ m⁻². RF power is 0.2 W.

| Pressure (Pa)/levels | Hump       | Dip        |
|----------------------|------------|------------|
|                      | 1s₅  | 1s₃  | 1s₂  | 1s₄  | 1s₅  | 1s₃  | 1s₂  | 1s₄  |
| 15                   | 11.1 | 0.78 | 2.4  | 2.1  | 4.7  | 0.50 | 1.1  | 0.99 |
| 30                   | 12.5 | 0.98 | 3.0  | 2.1  | 0.67 | 0.23 | 0.36 | 0.08 |
| 60                   | 16.6 | 1.1  | 3.7  | 2.7  | 1.1  | 0.19 | 0.45 | 0.09 |

(figure 6(b)). The upper glow region appears to be influenced much more weakly. The axial profiles of the intensities of the spectral lines support the effect observed with the imaging (figure 6(c)). In addition, they exhibit a slight increase of the intensity in the upper glow region. The maxima of intensity profiles in the bottom glow region shift several mm toward the center of the discharge in the presence of a microparticle cloud. Therefore, it is clear that a cloud of microparticles levitating in our plasma produces significant non-local influence on the discharge.
Figure 6. Images of the plasma glow at 30 Pa (white horizontal lines represent the electrodes): (a) microparticle-free discharge, (b) discharge with a cloud of microparticles. (c) Axial profile of the intensities of the spectral lines. Solid curve corresponds to the microparticle-free plasma and dashed curve to the plasma with a cloud of microparticles. The main effect of the microparticles on the glow is the extension of the bottom glow region into the central area of the discharge.

Figure 7. (a) Typical image of a discharge with a cloud of microparticles levitating in it (30 Pa). The cloud consists of a main thick cloud and a much smaller cloud, levitating above the main one. Attenuation $K_{ij}$ is taken into account only for the main cloud. (b) Comparison of the number densities of 1s$_2$ and 1s$_5$ states, determined with measured $K_{ij}^d$ (solid curve) and $K_{ij}^d = 1$ (dashed curve). Not accounting for the extinction of the plasma light by microparticles may lead to up to 25% overestimation of the number densities of states.

According to our method, for the number densities of the argon states to be determined in the presence of a microparticle cloud, attenuation $K_{ij}^d$ (figure 3) must be taken into account. There are two issues that alter the exact relevance of values of $K_{ij}^d$ estimated in section 2.3 to the actual values of the extinction in the self-absorption experiments. Firstly, Mie-scattering measurements are sensitive to geometrical factors like, e.g. view angle of the detector or angular composition of the incident light [23]. In a very simple estimation of $K_{ij}^d$ we were not able to
reproduce the geometry of the light source: for the self-absorption measurements we used a large mirror, which covered almost the entire rectangular viewport (80 mm height and 95 mm width), whereas to estimate \( K^d_{ij} \) we employed a cylindrically shaped Ar lamp, which was oriented vertically and covered the entire height of the viewport, but was only 6 mm thick in the horizontal direction. Secondly, as already mentioned, \( K^d_{ij} \) was estimated for a structure of microparticles different from (but nevertheless similar to) the one used in the self-absorption experiment. We, therefore, cannot depend on the high precision of the extinction correction of the number density of the states. In spite of this, we would like to demonstrate the importance of accounting for the microparticle-caused light attenuation.

We should also note that \( K^d_{ij} \) should certainly be different at different heights. In our consideration, we skip the axial dependence and simply use the values of \( K^d_{ij} \) from figure 3 for every point inside the ‘main’ microparticle cloud. We do not account for the microparticle-caused attenuation in a small cloud, located slightly above the main one, due to its smaller radial extension. We checked that for such small clouds attenuation appears to be undetectable. In figure 7(b), the results of the determination of \( n_j \) with measured \( K^d_{ij} \) and \( K^d_{ij} = 1 \) are compared. Neglecting the microparticle-caused attenuation under our conditions may lead to up to 25% overestimation of the number densities. This overestimation may be even larger in the experiments, where the structures of microparticles are denser and/or more extensive, e.g. under microgravity conditions [13] or when thermophoresis is used to compensate for gravity [24].

The axial distribution of the number density of states undergoes practically the same changes as the distribution of line intensities does in the presence of microparticles. The bottom hump of the distribution is stretched toward the midplane of the discharge (figure 8). Therefore, the strongest relative changes (double and more with respect to the microparticle-free plasma) are in the center of the discharge. This effect is much less pronounced at 15 Pa than at 30 and 60 Pa (figure 9). The upper hump remains practically unaffected by the presence of microparticles.

4. Discussion

4.1. Ionization balance

As already mentioned, microparticles suspended in a plasma volume may constitute a significant bulk loss of plasma particles. Let us try to estimate this loss for the conditions of our experiment. For this we have to compare the loss of the electrons on the microparticles with their production rate in a microparticle-free plasma. The axial distribution of electron temperature in \( \alpha \) RF discharges is significantly non-uniform [10]. It determines the observed two-humped distribution of brightness. The distribution of ionization should also reproduce this structure. The microparticle cloud in our experiment roughly occupies the volume, corresponding to one of these maxima. The second maximum is practically not affected by the presence of microparticles. This suggests the diffusive interchange between the two maxima is weak, allowing therefore to treat the ionization balance in them independently from each other.

We consider here three mechanisms of ionization: electron impact on metastable argon atom (\( 1s_3 \)), metastable pooling and electron impact on ground state argon atoms. We use the expressions for constants of these processes from [25] to calculate the respective rates for electron temperatures \( T_e = 1–7 \text{ eV} \) and electron densities \( n_e = 10^{14}–10^{15} \text{ m}^{-3} \). Line-of-sight averaged densities from table 1 with \( l \) equal to the electrode diameter (0.06 m) were used to
obtain the number densities of metastables. Electron impact ionization from the ground state dominated at $T_e \gtrsim 2 \text{ eV}$.

The loss of electrons on the microparticles is an essential function of their surface potential, which is in turn self-consistently connected to the conditions in the background plasma. We cannot calculate the electron loss self-consistently. Instead, we estimate the electron flux on microparticles with a certain fixed potential, immersed into a microparticle-free plasma, and compare it with the respective ionization rate.

The absolute value of the potential may lie between zero and the potential of an isolated particle, given, e.g. by orbital-motion-limited (OML) theory [27]. The OML electron flux is given by the following equation:

$$j_e(\phi) = \pi a^2 n_e \sqrt{\frac{8}{\pi m}} \frac{T_e}{T_e} \exp \left( \frac{\phi}{T_e} \right).$$

where $a$ is the radius of a spherical microparticle, $m$ is the electron mass and $\phi$ is the surface potential of a microparticle. We will consider the fluxes in the range $j_e(0) > j_e(\phi_{is})$, where $\phi_{is}$ is the surface potential of an isolated microparticle: $\sqrt{T_e/m} \exp(\phi_{is}/T_e) = \sqrt{T/M}(1 - \phi_{is}/T)$. 

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Figure 9. Pressure dependence of the effect of the cloud of microparticles on the number density of 1s5 level. Blue vertical lines represent the upper and lower boundaries of the main microparticle cloud. The effect of the presence of microparticles on the number densities of other states has a similar pressure dependence.

The bulk loss of electrons is then \( n_{d}j_{e}(\phi) \), where \( n_{d} \) is the density of microparticles, estimated for all the experiments to be of the order of \( 10^{11} \text{ m}^{-3} \).

On a map in figure 10, the \( (T_{e}, n_{e}) \) plane is divided into three regions (30 Pa)—where ionization rate exceeds both losses \( n_{d}j_{e}(\phi) \) and \( n_{d}j_{e}(0) \), where it lies in between them and where it is below both of them. The map looks qualitatively similar for other pressures. Being deep in the red region would mean that the presence of the microparticle cloud would not significantly affect the ionization balance. In the blue region, the plasma cannot be sustained due to the loss of electrons on suspended microparticles. The green region represents the transient situation. A possible situation in the green region is e.g. the following: the loss of electrons on microparticles is significant during their charging, but becomes much less significant after they are charged to equilibrium.

This treatment does not take into account depletion of electrons in the presence of microparticles, often associated with the so-called Havnes parameter [26]. This depletion would make sustainment of the plasma at a given \( T_{e} \) even more difficult.

As follows from figure 10, in the reasonable range of \( n_{e} \) and \( T_{e} \) for our plasma we can expect a strong effect of the microparticles on the ionization balance. The transformation that our plasma undergoes may be qualitatively understood in the following way. The line intensities
Figure 10. Color map presenting the qualitative comparison of ionization rate and loss of electrons on the microparticles for 30 Pa. In the red area, the ionization rate exceeds both \( n_d j_e(0) \) and \( n_d j_e(\phi_n) \), in the green area it is in between them and in the blue area loss on microparticles dominates over the ionization. In the reasonable range of parameters the region with a significant impact of microparticles on the ionization balance is found.

we observe are strong functions of the electron temperature. Since injection of microparticles does not at least increase the amplitude of the discharge brightness, the amplitude of the electron temperature does not seem to increase. Instead, the region with high \( T_e \) is spread quite far outside the main microparticle cloud so that diffusive influx of electrons into the cloud could compensate for the loss of electrons on the microparticles. This transformation, however, requires further experimental and theoretical investigations to reveal the mechanisms underlying it.

4.2. Metastables

Lifetimes of argon metastable states significantly exceed their typical diffusion times in plasmas. Therefore, metastable atoms can reach the surface of a microparticle levitating in a plasma and be quenched on it well before they decay. This aspect of metastable–microparticle interactions has been studied in detail in [12], where a very local effect of microparticles on the density of \( 1s_3 \) neon metastables was observed: the density decreased only within the dimensions of the microparticle cloud and was not perturbed outside. This is in contrast with the results of our measurements: we do not observe a significant change of the number density of metastable states inside the cloud of microparticles, instead we have a dramatic increase of it closer to the middle plane of the discharge.

Absorption of metastables in our case may be obscured by the disturbance of the ionization balance, discussed above. In [12], the plasma volume was much bigger: 13 cm RF-electrode diameter in the center of a 40 cm diameter chamber. Therefore, microparticles occupy a significantly smaller part of the discharge volume. In addition, in [12], significantly lower pressure (1–10 Pa) and significantly higher RF powers are used, resulting in spatial...
enlargement of the discharge features. Consequently, the vertical extension of the microparticle cloud becomes significantly smaller than the area with enhanced ionization. This does not allow microparticles to significantly influence the ionization balance in the case of \([12]\). A demonstration of the transition from the local to non-local influence of microparticles is also an interesting task for future investigations.

4.3. Surface of a microparticle as a source of species

The surface of a microparticle is not only a sink of plasma particles. It may also serve as a source of either impurities due to sputtering or of electrons due to secondary emission. It is known that under static pressure conditions in the PK-3 Nefedov chamber, nanoparticles grow due to the sputtering of either the melamineformaldehyde microspheres, lying on the bottom electrode, or those levitating in the volume of the chamber under microgravity conditions \([14]\). In this case, the amount of radicals in the discharge volume is fairly large and nanoparticles are grown on the timescale of minutes at, however, much higher RF power compared to that we use. Since in our experiments the gas in the chamber is refreshed every 40 s, we may most likely neglect the influence of sputtering on the plasma. Monitoring the emission spectra in between two gas pulses showed no dependence of their evolution on the presence of microparticles in the discharge. The periodic evolution of the spectra is therefore associated with periodic pressure variations only.

Secondary electron emission caused by ions and especially metastable atoms may become significant. For argon metastables the yield may approach unity on metallic surfaces \([29]\). Emission of electrons from surfaces under the effect of metastable rare-gas atoms is used in surface analysis (so-called metastable deexcitation spectroscopy, see e.g. \([30]\)) and takes place therefore on materials of different nature. To reveal the role of the secondary electron emission from the surfaces of microparticles levitating in plasmas, special dedicated experiments are required.

5. Conclusion

Microparticles, immersed in a RF-discharge plasma under gravity conditions levitate above the powered electrode in a glow region, i.e. an area with increased electron temperature. For the PK-3+ setup it was demonstrated that for large clouds of microparticles, whose axial extension is comparable to the axial extension of the glow region, their influence on the plasma is non-local. The presence of such a microparticle cloud leads to the enlargement of the bottom glow region, containing the microparticles. Measurements of the densities of 1s excited states of argon did not reveal significant changes in the amplitudes of their variation, whereas some local values increased by a factor of two or more in the presence of microparticles. The upper glow region stayed practically unperturbed. The most probable mechanism responsible for this influence is disturbance of the ionization balance by the levitating microparticles.

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References

[1] Bouchoule A 1999 Technological impacts of dusty plasmas *Plasmas: Physics, Chemistry and Technological Impacts in Plasma Processing* ed A Bouchoule (New York: Wiley)
[2] Vladimirov S V and Ostrikov K 2004 *Phys. Rep.* 393 175
[3] Fortov V E, Khrapak A G, Khrapak S A, Molotkov V I and Petrov O F 2004 *Phys.-Usp.* 47 447
[4] Dimoff K and Sym P R 1970 *Phys. Lett. A* 32 13
[5] Bouchoule A and Boufendi L 1993 *Plasma Sources Sci. Technol.* 2 204
[6] Bouchoule A and Boufendi L 1994 *Plasma Sources Sci. Technol.* 3 292
[7] Tachibana K, Hayashi Y, Okuno T and Tatsuta T 1994 *Plasma Sources Sci. Technol.* 3 314
[8] Böhm C and Perrin J 1991 *J. Phys. D: Appl. Phys.* 24 865
[9] Fridman A A, Boufendi L, Hibid T, Potapkin B V and Bouchoule A 1996 *J. Appl. Phys.* 79 1303
[10] Raizer Yu P, Shneider M N and Yatsenko N A 1995 *Radio-Frequency Capacitive Discharges* (New York: CRC Press)
[11] Denysenko I, Berndt J, Kovachevich E, Stefanovic I, Selenin V and Winter J 2006 *Phys. Plasmas* 13 073507
[12] Do H T, Kersten H and Hippler R 2008 *New J. Phys.* 10 053010
[13] Thomas H M et al 2008 *New J. Phys.* 10 033036
[14] Mikikian M et al 2003 *New J. Phys.* 5 19
[15] Mikikian M, Couëdel L, Cavarroc M, Tessier Y and Boufendi L 2007 *New J. Phys.* 9 268
[16] Ochkin V N 2009 *Spectroscopy of Low Temperature Plasma* (Berlin: Wiley)
[17] Gavare Z, Gött D, Pipa A V, Röpcke J and Skudra A 2006 *Plasma Sources Sci. Technol.* 15 391
[18] Klined worth M, Arp O and Piel A 2007 *Rev. Sci. Instrum.* 78 033502
[19] Takahashi K, Thomas H M, Morfill G E, Iblev A V, Hayashi Y and Adachi S 2008 Diagnosis in complex plasmas for microgravity experiments (PK-3 plus) *Multifacets of Dusty Plasmas* (AIP Conf. Proc. vol 1041) ed J T Mendoça, D P Resendes and P K Shukla (Berlin: Springer)
[20] Copley G H and Camm D M 1974 *J. Quant. Spectrosc. Radiat. Transfer* 14 899
[21] Aeschliman D P, Hill R A and Evans D L 1976 *Phys. Rev. A* 14 1421
[22] Schulze M, Yanguas-Gil A, von Keudell A and Awakowitz P 2008 *J. Phys. D: Appl. Phys.* 41 065206
[23] Nefedov A P, Petrov O F and Vaulina O S 1997 *Appl. Opt.* 36 1357
[24] Rothermel H, Hagl T, Morfill G E, Thoma M H and Thomas H M 2002 *Phys. Rev. Lett.* 89 175001
[25] Gudmundsson J T and Thorsteinsson E G 2007 *Plasma Sources Sci. Technol.* 16 399
[26] Melandsø F and Havnes O 1991 *J. Geophys. Res.* 96 5837
[27] Mott-Smith H M and Langmuir I 1926 *Phys. Rev.* 28 727
[28] Stoffels WW, Stoffels E, Swinkels G H P M, Boufnichel M and Kroesen G M W 1999 *Phys. Rev.* E 59 2302
[29] Dunning F B and Smith A C H 1971 *J. Phys. B: At. Mol. Phys.* 4 1696
[30] Sesselman W, Konrad H, Ertl G, Küppers J, Woratschek B and Haberland H 1983 *Phys. Rev. Lett.* 50 446