Crystal–liquid phase transitions in three-dimensional complex plasma under microgravity conditions

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Abstract. Complex (dusty) plasmas are composed of weakly ionized gas and charged microparticles and represent the plasma state of soft matter. Due to the “heavy” component, microparticles, and the low density of the surrounding medium, the rarefied gas and plasma, it is necessary to perform experiments under microgravity conditions to cover a broad range of experimental parameters which are not available on ground. The investigations have been performed onboard the International Space Station with the help of the PK-3 Plus laboratory. This laboratory was mainly built to investigate the crystalline state of complex plasma, the so-called plasma crystal, its phase transitions and processes in multi-particle mixtures. Due to the manipulation of the interaction potential between the microparticles it is possible to initiate a phase transition from isotropic plasma into electrorheological plasma. The crystal–liquid phase transition was obtained in large three-dimensional isotropic dusty plasma system. First observations of a transition of the dusty plasma system state due to variations of the plasma component density are presented.

1. Introduction
Complex plasmas are plasmas containing small solid particles, typically in the micrometer range, the so-called microparticles. These are dusty plasmas which are specially prepared to study fundamental processes in the strong coupling regime on the most fundamental (kinetic) level, through the observation of individual microparticles and their interactions. Many interesting phenomena can be studied starting from small two-dimensional (2D) and three-dimensional (3D) clusters [1], to larger 2D and 3D systems where collective effects play a dominant role [2].

In laboratory conditions, the microparticles are heavily affected by the force of gravity. Gravity leads to the sedimentation of the particles and can be balanced either by a strong electric field in the sheath of a discharge or through thermophoretic force due to a constant temperature gradient over the microparticle cloud. Additionally, there exist weaker forces like the neutral and ion drag forces, which nevertheless can play a very important role in the total force balance.

Under microgravity conditions, for example on the International Space Station (ISS), gravity is negligible. Therefore, the particles are pushed out of the strong electric field region close to the electrodes due to their negative charge and can form large, more or less, homogenous
particle clouds in the bulk of the discharge. Under these conditions, weaker forces like the ion drag force and the interparticle interactions become important and often dominate the motion and structure formation in complex plasma.

Since 2001 complex plasma research under microgravity conditions is continuously performed in a Russian–German cooperation onboard the ISS with the long-term laboratories PKE-Nefedov and PK-3 Plus, operational from 2001 to 2005 and from 2006 to 2013, respectively. The first laboratory, PKE-Nefedov, has already provided great insight into the behavior of complex plasma under microgravity conditions [3–6]. Fundamental investigations, like the formation of a particle free void in the center of the microparticle cloud induced by the ion drag force (underestimated by the state of the art theory, which has been improved by the adaption to the new experimental results [7]), or the first observation of bcc structures in plasma crystals [3] are only a few of the interesting results obtained there. The follow-up laboratory, PK-3 Plus, has been improved considerably compared to the first one and has been equipped with new diagnostic tools [8–10]. It has provided the next important step in research of complex plasmas under microgravity conditions. The obtained results are unique and opened up a new interdisciplinary research directions, in particular, related to the field of soft condensed matter. The purpose of this paper is to present some new results obtained using the PK-3 Plus laboratory onboard ISS.

2. Short description of PK-3 Plus laboratory

The PK-3 Plus setup has a well-balanced symmetrically driven rf-electrode system which provides a homogeneous distribution of the plasma with identical sheaths near both electrodes. This is necessary for a homogeneous distribution of the microparticles under microgravity conditions. A cross-sectional and perspective schematic of the microgravity setup, the PK-3 Plus chamber, is shown in figure 1 [8]. The vacuum chamber consists of a glass cuvette of form of a cuboid with a quadratic cross section. Top and bottom flanges are metal plates. They include the rf-electrodes, electrical feedthroughs and the vacuum connections. The electrodes are circular plates made from Aluminum with a diameter of 6 cm. The distance between the electrodes is 3 cm. The electrodes are surrounded by a 1.5 cm wide ground shield including three microparticle dispensers on each side. The dispensers are magnetically driven pistons with a storage volume at their ends. The storage volumes are filled with microparticles and covered with a sieve with an adapted mesh size. The microparticles are dispersed through the sieve into the plasma chamber by electromagnetically driven strokes of the piston.
Figure 2. The transition from an isotropic fluid (a) to a string fluid (b) observed in the PK-3 Plus laboratory onboard the ISS. Particles of 6.8 $\mu$m, argon pressure is 10 Pa, interparticle distance is 370 $\mu$m. Strings are apart from each other at 580 $\mu$m. Snapshots are obtained by the high-resolution camera.

The optical particle detection system consists of laser diodes with cylindrical optics to produce a laser sheet perpendicular to the electrode surface. Three progressive scan CCD-cameras observe the reflected light at 90° with three different magnifications and fields of view. An overview camera has a field of view of $58.6 \times 43.1$ mm$^2$. It shows the full field between the electrodes. A second camera has a field of view of $35.7 \times 26.0$ mm$^2$. It shows the left part of the interelectrode system (about half of the full system). A third camera with the highest resolution can be moved along the central axis and has a field of view of $8.1 \times 5.9$ mm$^2$. The latter is used for high precision position measurements of the microparticles. The cameras and lasers are mounted on a horizontal translation stage allowing a depth scan through and, therefore, a 3D view of the complex plasma [11].

3. Results on phase transitions in 3D complex plasmas

3.1. Electrorheological effect in complex plasmas

One of the important results achieved using the PK-3 Plus laboratory was the first observation of the electrorheological effect in complex plasmas or a phase transition from an isotropic fluid into a so-called electrorheological string fluid [12].

The formation of such string fluids, or general electrorheological plasmas, is possible due to the manipulation of the interaction potential between the microparticles along the field line. It can be changed from an isotropic screened Coulomb to an asymmetric attractive potential through accelerating ions by an additional ac voltages applied to the electrodes at frequencies above the dust plasma frequency. The ions then produce wake regions above and below the particles along the electric field axis, while the particles cannot respond. It has been shown [12] that the effective interparticle interaction in this case is determined by the time-averaged wake potential. The field-induced interactions in dusty plasmas are identical to interactions in conventional electrorheological fluids with dipoles $d = 0.65 Q \lambda \nu_{\text{ion}} / \nu_{\text{th}}$, where $Q$ is the particle charge, $\lambda$ is the ion screening length, $\nu_{\text{ion}}$ is the ion drift velocity and $\nu_{\text{th}}$ is the ion thermal velocity.

In experiments, the ac voltage at a frequency of 100 Hz was applied to the rf electrodes with the amplitude voltage between 26.6 and 65.6 V varied in steps of 2.2 V. At weak voltages charged particles formed a strongly coupled isotropic fluid phase. As the voltage was increased above a certain threshold, particles rearranged themselves and became more and more ordered, until eventually well-defined particle strings were formed along the direction of the field. In these experiments we used microparticles of different diameters (1.55, 6.8 and 14.9 $\mu$m) and Ar gas at pressures between 8 and 15 Pa. The example of the transition to the electrorheological plasma state is presented in figure 2. The transition between the isotropic fluid phase and
Figure 3. Freezing indicator $R$ (the Raveche–Mountain–Streett ratio) for complex plasmas composed of small (blue triangles) and large (red circles) particles.

the electrorheological state was fully reversible—decreasing the field brought the particles back into their initial isotropic state. The trend to form strings increased with a particle size, in agreement with theoretical estimates. The molecular dynamic simulations performed with similar parameters yielded remarkable agreement with the experiments.

3.2. Phase transition by manipulating the neutral gas pressure

We performed experimental investigations of the fluid-solid phase transitions in large 3D complex plasmas under microgravity conditions. These phase changes were driven by manipulating the neutral gas pressure. Detailed analysis of complex plasma structural properties allowed us to quantify the extent of ordering and accurately determine the phase state of the system. Evaluation of various freezing and melting indicators gave further confidence regarding the phase states. It was observed that the system of charged particles can exhibit melting upon increasing the gas pressure, in contrast to the situation in ground-based experiments where plasma crystals normally melt upon reducing the pressure [13]. This illustrates important differences between generic (e.g. similar to conventional substances) and plasma-specific mechanisms of phase transitions in complex plasmas.

The experiments have been carried out in argon at a low rf-power [14]. We used two different sorts of particles in the two distinct experimental runs: SiO$_2$ spheres with a diameter 1.55 $\mu$m and melamine formaldehyde spheres with a diameter 2.55 $\mu$m. The experimental procedure, identical in these two runs, was as follows. When the particles formed a stable cloud in the bulk plasma, the solenoid valve to the vacuum pump was opened, which resulted in a slow decrease of the gas pressure $p$. Then, the valve was closed and the pressure slowly increased due to the gas streaming in. During the pressure manipulation ($\approx$ 6 minutes in total), the structure of the particle cloud was observed. The observations covered the pressure range from $p \approx 15$ Pa, down to the lowest pressure of $p \approx 11$ Pa and then up to $p \approx 21$ Pa.

In order to get three-dimensional particle coordinates, 30 scans were performed. Scanning was implemented by simultaneously moving laser and cameras in the direction perpendicular to the field of view with the velocity 0.6 mm/s. Each scan takes $\approx 8$ s, resulting in the scanning depth of $\approx 4.8$ mm; the interval between consecutive scans is $\approx 4$ s. The particle positions were then identified by tomographic reconstruction of the 3D-pictures taken with the high resolution camera observing a region $8 \times 6$ mm$^2$, slightly above the discharge center.

To characterize a structural state of the dusty plasma systems observed we shall use as an example the Raveche–Mountain–Streett criterion of freezing [15], which is based on the properties of the radial distribution function $g(r)$ in the fluid phase. It states that near freezing, the ratio of the values of $g(r)$ corresponding to its first nonzero minimum and to the first maximum, $R = g(r_{\text{min}})/g(r_{\text{max}})$, is approximately constant, $R \approx 0.2$. This criterion describes fairly well freezing of the classical Lennard-Jones fluid, but is not truly universal (i.e., the ratio...
Figure 4. Video images of the restored dusty plasma system for the argon pressure 20 Pa. (a) $U_{\text{eff}} = 19.0$ V; (b) $U_{\text{eff}} = 16.3$ V; (c) $U_{\text{eff}} = 13.1$ V.

Figure 5. Video images of the restored dusty plasma system for the argon pressure 40 Pa. (a) $U_{\text{eff}} = 16.2$ V; (b) $U_{\text{eff}} = 14.5$ V; (c) $U_{\text{eff}} = 12.5$ V.

$R$ can somewhat vary for different systems. Figure 3 shows the calculated values of the freezing indicator $R$ for different scans. Applying the threshold condition $R \simeq 0.2$ would imply that the system of small particles melts upon an increase in the neutral gas pressure (second half of the observation sequence), while the system of large particles remains in the solid state. This is consistent with the results of more detailed structural analysis [14].

3.3. Phase transition by changing the plasma component density

Several experiments have been performed to study peculiarities of the crystal-liquid phase transition in the 3D dusty plasma system when varying the voltage between electrodes of the plasma chamber. The change of the interelectrode voltage leads to a change of the plasma component density and one of the important parameters determining a behavior of the dusty plasma system, namely, a screening length. In the experiments monodisperse SiO$_2$ particles of 1.55 $\mu$m diameter were used.

The procedure of the experiments was the following one. The radio-frequency discharge was generated at the given argon pressure and at the given interelectrode voltage. Then a formation of an ordered structure took place. After the structure formation by applying for a short time a low frequency voltage (frequency of 255 Hz, amplitude of voltage 13 V) from the functional generator the structure was partially destroyed and the process of restoring the initial ordered structure was registered. During the restoring process a scanning of the structure at a depth from minus 4.8 mm up to plus 4.8 mm was performed. The velocity of the scanning was 0.6 mm/s. Figure 4 presents video images of the dusty plasma system restored 3 minutes after the destruction. The images were obtained by the high resolution camera at the argon pressure
20 Pa at different values of the interelectrode voltage ($U_{\text{eff}} = 19.0, 16.3$ and 13.1 V respectively). It is clearly seen a decrease of the ordering level when rf voltage at the electrodes is decreasing, that is, when the ion density is decreasing.

Figure 5 presents video images of the restored dusty plasma system for the argon pressure 40 Pa. The data presented show that in this case the ordering level of the system is changing in the direction opposite in comparison with the previous case.

To interpret the results obtained we made evaluations of the parameters of the dusty plasma systems observed. Figures 6 and 7 show the pair correlation functions determined on the results of a scanning of the each system. The experimental correlation functions give a possibility to evaluate a level of the dusty plasma system ordering and to determine an interparticle distance.

For the analysis of the dusty plasma system state we use the Raveche–Mountain–Streett criterion mentioned earlier. Note that in the case when $R$ exceeds 0.2 the dusty plasma system is in the fluid state and in the case when $R$ is less than 0.2 the system is in the well ordered (crystal) state.
Figure 8. Change of the criterion $R$ on the interelectrode voltage at different pressure.

Figure 8 shows a change of the criterion $R$ when the interelectrode voltage changes, that is, the plasma component density changes. It is seen that a character of the change of the ordering at two different values of the argon pressure has quite an opposite behavior. At the argon pressure 20 Pa an ordering of the system decreases when there is a reduction of the voltage $U_{\text{eff}}$: the dusty plasma system melts at a reduction of the plasma component density. In the case of the argon pressure 40 Pa when the interelectrode voltage increases the dusty plasma system ordering increases as well.

To obtain values of the plasma component density (in the first turn, the ion density) we use the results of calculations on the base of the SIGLO-2D model [8]. The evaluation of plasma parameters was made without taking into account dust particles. The obtained ion densities for pressures 20 and 40 Pa are presented in tables 1 and 2. In the tables there are also the values of the screening length $\lambda$ and the ratio of the screening length to the ion mean free path $\lambda/l_i$ (this ratio is often called the ion collisionality index). In our case the screening length when the electron density substantially (by two orders of a value) exceeds the ion temperature equal the room temperature $T_i = 300$ K is determined by the ion Debye length. The ion mean free path was determined on the cross section of the argon ions overcharging. At the pressure 20 Pa $l_i$ is equal to 196 $\mu$m and at the pressure 40 Pa it is equal to 98 $\mu$m. The tables contain also values of the interparticle distances and the screening parameter $k$, where $k = \Delta/\lambda$, $\Delta$ is interparticle distance. The values of the exponential multiplier $e^{-k}$ in the coupling parameter $\Gamma$ ($\Gamma = (Q^2/Tp\Delta)e^{-k}$, where $Q$ is microparticle charge, $T_p$ is kinetic temperature of dust particles) are presented in the tables as well.

As it is seen from the data given in the tables the value of the exponential multiplier increases when the ion density decreases. This is true for both argon pressures (20 and 40 Pa). Therefore, the different character of a change of the dusty plasma system ordering upon the ion density, apparently, it is connected with a difference in changing a charge of dust particles when the interelectrode voltage changes. Let us make some qualitative evaluations using results of the theoretical studies. It is known from the theoretical studies [1] that a dust particle charge is a function of the ion collisionality index $\lambda/l_i$. In the range of respectively low values of this index a dust particle charge decreases with an increase of $\lambda/l_i$. The values of $\lambda/l_i$ for the argon pressure
Table 1. Parameters of the dusty plasma system at $P = 20$ Pa.

| $U_{\text{eff}}$ (V) | $n_i$ (cm$^{-3}$) | $\lambda$ (µm) | $\lambda/l_i$ | $\Delta$ (µm) | $k$ | $e^{-k}$ | $Q$ (e) | $U$ (eV) |
|----------------------|------------------|----------------|---------------|---------------|-----|---------|---------|--------|
| 19.0                 | 8 $\times$ 10$^8$ | 43             | 0.22          | 131           | 3.05| 0.047   | 1.6 $\times$ 10$^3$ | 1.34   |
| 13.1                 | 5 $\times$ 10$^8$ | 54             | 0.28          | 122           | 2.26| 0.1     | 8.8 $\times$ 10$^2$  | 0.96   |

Table 2. Parameters of the dusty plasma system at $P = 40$ Pa.

| $U_{\text{eff}}$ (V) | $n_i$ (cm$^{-3}$) | $\lambda$ (µm) | $\lambda/l_i$ | $\Delta$ (µm) | $k$ | $e^{-k}$ | $Q$ (e) | $U$ (eV) |
|----------------------|------------------|----------------|---------------|---------------|-----|---------|---------|--------|
| 16.2                 | 9.0 $\times$ 10$^8$ | 40             | 0.4           | 136           | 3.4 | 0.033   | 1.9 $\times$ 10$^3$ | 1.28   |
| 12.5                 | 6.5 $\times$ 10$^8$ | 47.5           | 0.49          | 136           | 2.86| 0.057   | 1.4 $\times$ 10$^3$  | 1.19   |

20 Pa correspond to the mentioned range. Since the coupling parameter $\Gamma$ depends on $Q^2$ the decrease of the particle charge at 20 Pa, we suppose, prevails over an increase of the exponential multiplier $e^{-k}$ and a decrease of a level of the dusty plasma system takes place.

When the argon pressure increases the ion collisionality index increases as well. It is known [1] that at such a noticeable increase of this index the dependence of the particle charge on the index $\lambda/l_i$ is shifted to the region of the minimum (or a slight dependence of the particle charge on the ion collisionality index). In this case it should be expected that the main input to the change of the coupling parameter $\Gamma$ follows from a change of the exponential multiplier $e^{-k}$. Note that this multiplier, as seen from the table 2 increases by 2 times. In connection with the mentioned remarks at the pressure 40 Pa we observe an increase of the dusty plasma system ordering (see figure 8).

4. Conclusion

The presented results show several examples of phase transitions in the 3D complex plasma system under microgravity conditions obtained with the help of the PK-3 Plus laboratory onboard the International Space Station. The manipulation of the interaction potential between the microparticles along the electric field line gives a possibility to obtain and to study a formation of the string fluid plasma system.

The manipulation of the neutral gas pressure allows researchers to observe the fluid-solid phase transitions in the large 3D complex plasma. The system of charged particles can exhibit melting upon increasing the gas pressure, in contrast to the situation in ground-based experiments where 2D plasma crystals normally melt upon reducing the pressure.

The other quite a new observation concerns a study of transitions of the dusty plasma system state due to variations of the ion density. It was demonstrated a rather complicated character of the dependence of the system order upon the ion density. In the investigated argon pressure range from 20 to 40 Pa we have observed the change of the phase transition direction. For the lower pressure the dusty plasma system demonstrates the transition from the crystal state to the less ordering system when the ion density decreases. For the greater pressure a character of the system order change is the opposite one: at the decrease of the ion density there is a transition from the liquid state to the well ordered system.
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References
[1] Fortov V E and Morfill G E (eds) 2010 Complex and Dusty Plasmas: From Laboratory to Space Series in Plasma Physics (Boca Raton, FL: CRC Press)
[2] Fortov V E, Ivlev A V, Khrapak S A, Khrapak A G and Morfill G E 2005 Phys. Rep. 421 1–103
[3] Nefedov A P et al 2003 New J. Phys. 5 33
[4] Lipaev A M et al 2007 Phys. Rev. Lett. 98 265006
[5] Samsonov D et al 2003 Phys. Rev. E 67 036404
[6] Khrapak S et al 2003 Phys. Plasmas 10 1–4
[7] Khrapak S A, Ivlev A V, Morfill G E and Thomas H M 2002 Phys. Rev. E 66 046414
[8] Thomas H M et al 2008 New J. Phys. 10 033036
[9] Molotkov V I, Thomas H M, Lipaev A M, Naumkin V N, Ivlev A V and Khrapak S A 2015 Int. J. Microgravity Sci. Appl. 32 320302
[10] Khrapak A G et al 2016 Contrib. Plasm. Phys. 56 253–62
[11] Naumkin V N, Zhukovitskii D I, Molotkov V I, Lipaev A M, Fortov V E, Thomas H M, Huber P and Morfill G E 2016 Phys. Rev. E 94 033204
[12] Ivlev A V et al 2008 Phys. Rev. Lett. 100 095003
[13] Thomas H M and Morfill G E 1996 Nature 379 806–9
[14] Khrapak S A et al 2012 Phys. Rev. E 85 066407
[15] Raveche H J, Mountain R D and Streett W B 1974 J. Chem. Phys. 61 1970–84