

Photophoretic force on microparticles in complex plasmas

C-R Du^{1,a}, V Nosenko², H M Thomas², A Müller³, A M Lipaev⁴, V I Molotkov⁴, V E Fortov⁴, and A V Ivlev^{3,b}

¹College of Science, Donghua University, Shanghai 201620, PR China

²Institut für Materialphysik im Weltraum, Deutsches Zentrum für Luft- und Raumfahrt (DLR), 82234 Weßling, Germany

³Max Planck Institute for Extraterrestrial Physics, Garching 85741, Germany

⁴Joint Institute for High Temperatures, Moscow 125412, Russia

E-mail: ^achengran.du@dhu.edu.cn, ^bivlev@mpe.mpg.de

Abstract. Experimental observations are presented of unusual (“abnormal”) microparticles, having trajectories very different from those of the majority of microparticles in the particle cloud in PK-3 Plus chamber (on board the International Space Station). To quantitatively study the mechanism driving this “irregular” motion, we performed a series of experiments with quasi-two-dimensional complex plasmas in a modified GEC rf reference cell in a ground-based laboratory. The results show that the average particle velocity increases with illumination laser power, particularly for the “abnormal” particles. We suggest that the photophoretic force provides an important contribution to the drive, and briefly discuss the mechanism leading to this effect. Optical microscopy results indicate that the “abnormal” particles could be those having deformations or defects on their surface.

1. Introduction

Complex plasmas are low-pressure gas discharge plasmas containing extra solid micron-sized particles [1]. Through the interaction with the plasma components (ions and electrons), the microparticles gain negative charges and interact with each other via screened Coulomb (Yukawa) interaction. The charge and the strength of the interaction depend on the particle size and plasma conditions. Being one of soft-matter states [2], complex plasmas exhibit macroscopic softness and have metastable states (crystal [3], glass [4] or liquids [5]) depending on external conditions. In the laboratory, particles can be illuminated by a laser sheet and individual particle motions can be recorded by a video camera for analysis. This gives us a unique opportunity to study generic processes in classical continuous media at an “atomistic” level [6].

Since the discovery of plasma crystals [3, 7, 8], plenty of fundamental physics has been studied experimentally in two-dimensional (2D) and three-dimensional (3D)

complex plasmas such as melting process [9, 10], diffusion [11, 12], heat transport [13, 14]. To study some phenomena such as viscosity [15] and lattice waves [11, 16] in 2D, it was sufficient to confine microparticles in a single layer and record the particle motion in the equilibrium state using video microscopy. However, in other experiments, external excitations were applied to heat the particle cluster [10] or to accelerate the particles in certain direction [17]. For example, to study recrystallization process of a plasma crystal, two wires were installed in the plasma chamber to provide an electric impulse so that the plasma crystal was melted instantaneously [18]. Another efficient external excitation is laser. Nosenko *et al* applied a laser beam to heat a strip of microparticles in a plasma crystal to study heat transport [10] and microstructure [19]. Melzer used a laser beam to excite collective modes in finite Coulomb clusters [20] through radiation pressure and thermophoresis. In these experiments, the power of the laser was relatively high and the direction of the force strongly depended on the relative position of microparticles to the orientation of the laser beam.

Recently, it was discovered that the direction of particle motion is not always correlated to the direction of the laser beam [21]. This phenomenon was also observed in the experiments under microgravity conditions [22]. It was suggested that this “irregular” motion was caused by the photophoretic force: When the surface of a particle is heated nonuniformly by light, the surrounding gas molecules rebound off its surface with different velocities creating a net force on the particle [23, 24, 25].

In this paper, we present a series of dedicated experiments to study photophoretic force on microparticles in complex plasmas. In section 2, we introduce experimental observations in PK-3 Plus laboratory on board the International Space Station (ISS). In section 3, we present a series of dedicated experiments performed in the laboratory to study the correlation of particle velocities on the illumination power. The mechanism of the photophoretic force is discussed in section 4. Finally, we provide a short conclusion in section 5.

2. Observations of “abnormal” particles under microgravity conditions

Through years of experiments in PK-3 Plus laboratory on board ISS, we observed on multiple occasions that single particles behaved quite differently from others in the particle cloud.

2.1. Experimental setup: PK-3 Plus Laboratory

The PK-3 Plus Laboratory was an experimental facility for the investigation of 3D complex plasmas under microgravity conditions on board the International Space Station. The setup consisted of a capacitively coupled radio-frequency (rf) chamber, advanced electronic components (including rf generator and low-frequency function generator) and gas-vacuum system for plasma discharge, optical diagnostic system for particle detection, and control system [26]. Six particle dispensers containing particles

of different materials and sizes were installed in the electrodes [27]. With the presence of plasma discharge under microgravity conditions, microparticles could be injected into the chamber and confined in the center by the plasma fields, forming a large 3D particle cloud. Due to the ion drag force, a “void” was often formed in the center of the particle cloud, see Fig. 1. The “void” could be closed by carefully tuning the discharge conditions, providing a much better homogeneity of the complex plasma. A complete description of PK-3 Plus laboratory can be found in the reference [26].

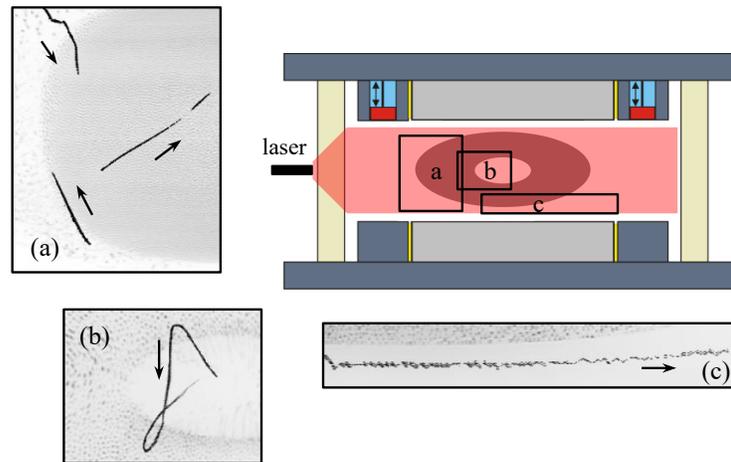


Figure 1. Observation of typical “abnormal” particle events in PK-3 Plus laboratory on board the ISS. A sketch of PK-3 Plus chamber is provided. Illumination laser sheet shined into the chamber through a quartz window. Three examples are selected and shown in panels (a), (b) and (c), corresponding to the three regions marked in the sketch. The images are superpositions of sequences of 77 consecutive video frames. The pixel intensity of the background is reduced to highlight the trajectories of “abnormal” particles. (a) “Abnormal” particles moved into, inside or at the periphery of the particle cloud ($U_{rf,eff} = 10$ V, $P = 30$ Pa); (b) An “abnormal” particle moved in a “ ∞ ” pattern in the void when a heart-beat instability was present [28] ($U_{rf,eff} = 8$ V, $P = 110$ Pa); (c) An “abnormal” particle moved outside of the particle cloud ($U_{rf,eff} = 14$ V, $P = 110$ Pa).

We applied video microscopy to diagnose the dynamics of individual particles. The microparticles are illuminated by a laser sheet shining through a view port of the chamber. The video camera, with its optical axis orthogonal to the laser sheet, records the laser light scattered off the particles at 90° . Thanks to the bandpass filter corresponding to the wavelength of the illumination laser, individual particles are registered as clusters of connected bright pixels and the background light is filtered out, leaving a clean black background. Both video cameras and laser diode are fixed on a platform and can move together to perform a scan with a given speed. This is particularly useful to visualize a 3D structure by scanning the whole volume of a complex plasma cloud.

In PK-3 Plus laboratory, three progressive scan PAL CCD cameras were installed to record particles at the plane of laser sheet with different fields of view. The recording rate can reach 50 frames per second (fps) [26, 27]. The recorded sequence of images can be analyzed by special software. Individual particle positions can be identified. By tracking consecutive positions of individual particles, particle velocity can be deduced.

2.2. Results

As the majority of the microparticles confined in the discharge exhibited collective dynamics, from time to time there were some particles which behaved differently. Often those particles had a different image shape and moved either with a much higher velocity or in a different direction comparing with the surrounding particles. The trajectories were rather random and they did not agree with those driven by plasma potential or ion drag force. We call those particles with “irregular” trajectories “abnormal” particles. Here, we show a few examples selected from various observations. As we see in figure 1(a), while most particles (SiO_2 particles with a diameter of $1.55 \mu\text{m}$) moved slowly in the cloud, an “abnormal” particle traveled across half of the cloud within a very short time (right). In the same panel, we can see another “abnormal” particle penetrate into the cloud from outside (left top) and a third particle move along the periphery of the cloud (left bottom). In another experiment, while the majority of the particles (melamine-formaldehyde particles with a diameter of $6.8 \mu\text{m}$) oscillated due to the heartbeat instability [29, 28], an “abnormal” particle moved independently back and forth across the void of the cloud, forming a “ ∞ ”-like trajectory, see figure 1(b). Sometimes we could even see a single “abnormal” particle move outside of the cloud [consisted of the same type of the particles in figure 1(b)] with a ultralong trajectory, rather independent of the particle cloud, see figure 1(c). In all these cases, the “abnormal” particles were “trapped” in the laser sheet and thus could be seen for rather long time. Such effect is known as “light tweezers” effect.

The effect became more evident in the scan experiments. In many experiments, as the camera and the laser moved together along the cameras’ line of sight (see the sketch in figure 1 in Ref. [27]), we observed that “abnormal” particles followed the laser sheet in the scan direction as in one typical example shown in figure 2. In this particular example, there were seven “abnormal” particles moving differently from surrounding particles within a movie of 2 seconds. Note that the laser sheet was moving in the z direction all the time and each layer in the 3D reconstruction in figure 2 was recorded at a different time. This is clearer if we look at figure 2(a-c). Here, we track the particle positions and plot them in three panels, corresponding to three orthogonal orientations. On one hand, all seven particles traveled at the same velocity (equal to laser scanning speed) in the z direction. On the other hand, they moved rather randomly in the y direction. Note that in the x direction, six out of seven particles tended to move in the positive direction, implying a dominant role of the radiation pressure force.

It is worthwhile mentioning that the “abnormal” particles were susceptible to

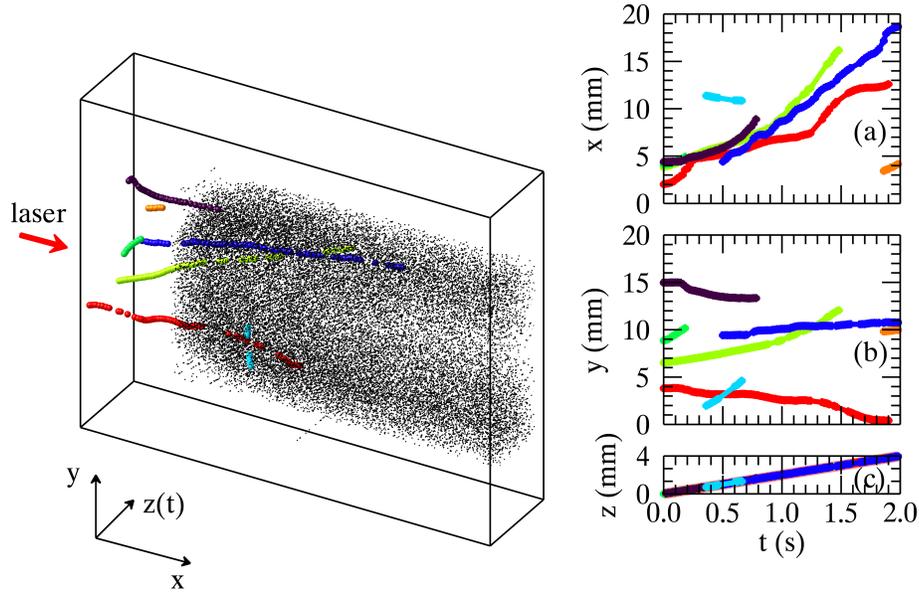


Figure 2. 3D trajectories of “abnormal” particles in PK-3 Plus laboratory on board the ISS (left). The cloud volume was scanned (2 mm/s) in the z direction by laser sheet and each layer was recorded in a frame at a time, see the supplemental material. The time t is represented by the z axis, and the trajectories of “abnormal” particles inside the cloud are obtained by 3D reconstruction. The “abnormal” particles follow the laser sheet in the z direction. These “abnormal” particles were tracked and their positions in all three components are plotted (right). Each single “abnormal” particle is marked by one color.

both radiation pressure and photophoretic force. This synergic effect was particularly noticeable in the longitudinal direction with respect to the laser beam direction, see figure 1(c) and figure 2. In the transverse direction, the major contribution was from the photophoretic force. Comparing the irregular trajectories of “abnormal” particles in both directions, the motion in transverse direction was as strong as in longitudinal direction, see figure 1(a,b) and some particle trajectories in figure 2. This signifies a strong contribution of the photophoretic force on the “abnormal” particles.

3. Dedicated studies of photophoretic force in ground laboratory

In order to study the effect of photophoretic force on the particle motion quantitatively, we performed a series of dedicated experiments in a modified Gaseous Electronics Conference (GEC) rf reference cell in a ground-based laboratory.

3.1. Experimental setup: GEC rf reference cell

GEC rf reference cell is a widely used rf plasma reactor suitable for studies of basic discharge phenomena [30]. For the purpose of investigation of 2D complex plasma, the top metal plate was replaced by a glass window, providing an additional view port [13, 19, 31].

In our experiments, argon plasma was sustained using a capacitively coupled rf discharge at 13.56 MHz. The input power was set at 20 W. We injected transparent[‡] monodisperse polystyrene (PS) particles into the chamber to create a 2D particle layer suspended above the bottom rf electrode. The particles have a diameter of $11.36 \pm 0.12 \mu\text{m}$, mass density of 1.05 g/cm^3 , and refractive index of 1.59. Gas pressure was maintained at about 0.65 Pa, corresponding to the damping rate $\gamma \simeq 0.91 \text{ s}^{-1}$ [32]. In addition, we placed two aluminium bars parallel to each other on the bottom electrode with a separation of 9 cm. They deformed the suspension to an elliptical shape and confined the fast moving particles in the vicinity of its periphery. By including the majority of the fast moving particles inside the field of view, we could significantly improve the statistics.

We also employed video microscopy for particle diagnostics in the GEC rf reference cell. To obtain better resolution, we used a megapixel video camera (Photron FASTCAM 1024 PCI) mounted above the chamber, capturing a top view with a size of $42 \times 42 \text{ mm}^2$. The recording rate was set at 60 fps [13, 31]. For fast moving particles, a cluster of connected bright pixels in one frame, representing a single particle, has an elongated shape. Taking advantage of this feature, we can deduce the velocity simply by dividing the length of the long axis in the cluster by the inverse frame rate, see the text below.

3.2. Results

In the experiments, particles were injected into the chamber in the presence of plasma. In order to get rid of the aggregates and other contaminants from the particle dispenser, we performed a “purification” procedure and removed heavy particles including aggregates [31]. However, this procedure could not remove the particles of small size or with defects on their surface, which in turn provided us the opportunity to study the laser effect on these “abnormal” particles. By scanning the laser vertically, we confirmed that these “abnormal” particles were above the main particle layer with a vertical distance smaller than $100 \mu\text{m}$ [31] (the width of the laser sheet).

Figure 3 shows a snapshot of the particle suspension. Half of the particle cloud was included in the view with a clear elliptical shape. As we see in the figure, the “abnormal” particles moved generally much faster than other particles in the layer, leaving a longer trajectory. Most of the time they moved freely at the periphery of the cloud, while occasionally they could also penetrate the cloud, heating the particle layer due to the

[‡] The illuminating laser light can generally reach any part of a transparent particle.

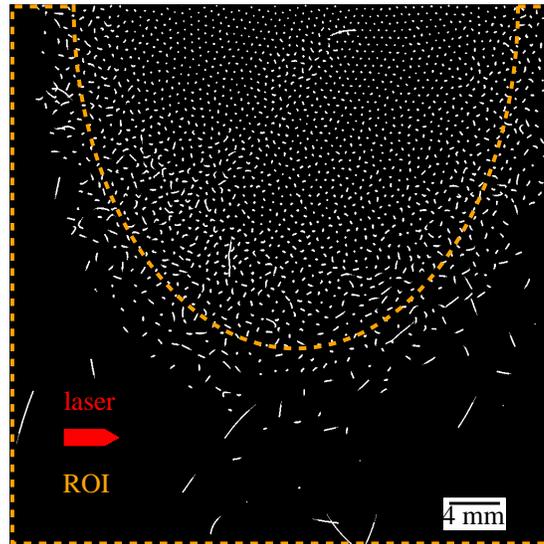


Figure 3. Snapshot of particles in the GEC chamber from the top view. The ROI is the periphery area of the particle cloud, enclosed by orange dashed curves. Particle velocities were deduced from the length of the trajectories. The direction of the laser sheet is marked by the red arrow.

ion wake in the sheath [31].

In order to study the photophoretic force on microparticles, we adjusted the laser power P_L to three different values (14 mW, 45 mW and 99 mW) and studied the dependence of particle velocity on laser power: The absolute values of the horizontal velocities were derived from the length of the trajectories with an accuracy of ≈ 2.5 mm/s (pixel size divided by the inverse frame rate), as demonstrated in figure 3. In the sheath the electric field together with the gravity imposes strong vertical confinement, and thus the vertical velocity cannot be directly measured. We separate the velocities in the longitudinal and transverse components with respect to the laser beam direction. Since some extra particles can strongly interact with the particles in the layer via non-reciprocal interaction [33], making the situation rather complicated, we merely focus on the periphery area, enclosed by the orange dashed curves in figure 3.

Figure 4 shows the dependence of the particle velocity distribution in the region of interest (ROI) on the laser power. The velocity distributions in the upper, middle, and lower panels were measured 10, 40 and 160 minutes after the dispensation of microparticles in the plasma, respectively. As we see in all six panels, the amount of the fast particles increases with the increase of laser power. This shows that indeed the photophoretic force provides the major contribution to the driving force for those extra particles. As we compare the left panels with the right panels, the velocity is

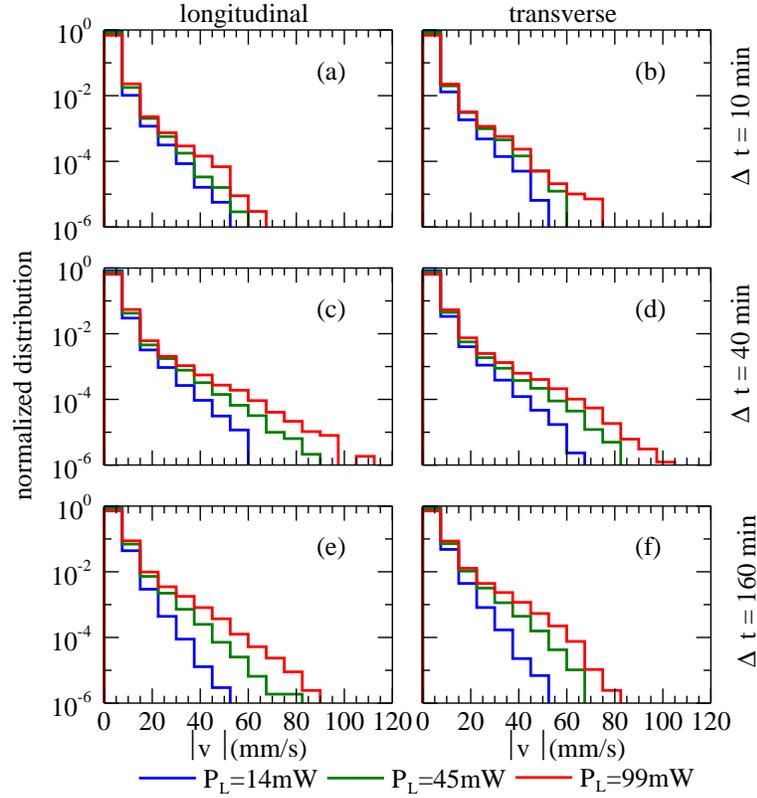


Figure 4. Measurement of the absolute value of the particle velocity distribution (normalized) in longitudinal (left panels) and transverse (right panels) direction with respect to the laser beam direction at different laser powers P_L in log scale. From top to bottom, the measurements were taken at $\Delta t = 10, 40$ and 160 minutes after dispensation of microparticles in the plasma.

weakly correlated with the laser beam direction \S . Furthermore, as we compare panels from top to bottom, the number of fast particles increases with time initially. It then reduces slightly after very long time. This might be explained by plasma processes such as etching of the particle material [34]. However, these plasma processes are beyond the scope of this paper.

4. Discussion

It was possible to collect particles in the modified GEC reference cell either prior or after the particles were dispensed in the plasma. We performed a survey on the particles' morphology using optical microscope. Standard PS particles are perfectly spherical and have a smooth surface, as shown in the top left panel in figure 5. The majority of the

\S In contrast to the experiments performed in PK-3 Plus laboratory, the effect of the radiation pressure is much less pronounced here, presumably due to a different material of microparticles and different experimental conditions.

collected particles fell in the category of standard particles. But there also existed a few deformed microparticles in the sample. As we see in the figure 5, the majority of the deformed particles were smaller than the standard particles, as expected. Based on the measurement of the diameter of our samples, the deformed particles could be about 20% smaller than the standard particles. Moreover, out of a comparable size of particle cloud, the number fraction of the deformed particles under microscope observation was about 1%, which roughly agreed with that of the “abnormal” particles recorded by video camera in the experiment.

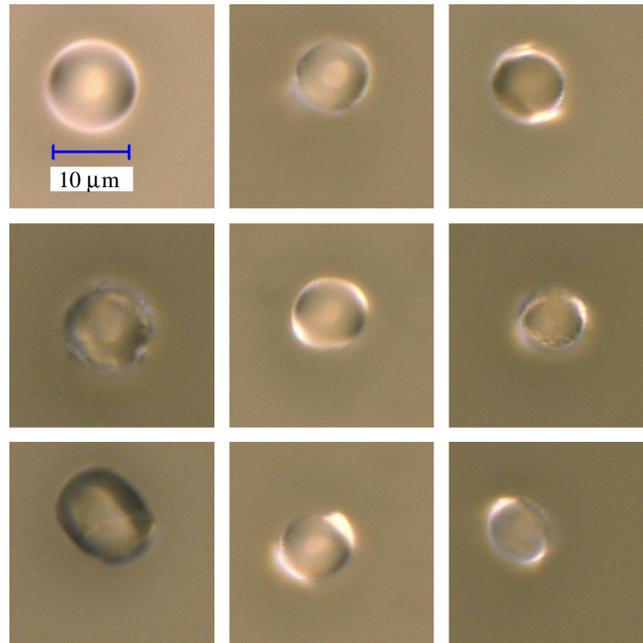


Figure 5. Morphology of microparticles under optical microscope. The top left panel is a standard PS particle with a diameter of $11.36 \mu\text{m}$. The rest of the panels show microparticles with deformations or defects on their surface.

Combining the study of the morphology of microparticles with particle dynamics in both experimental devices, we attribute the driving mechanism of those fast moving particles to the photophoretic force. Photophoresis occurs as a particle is non-uniformly heated by a beam of light. The surrounding gas atoms colliding with a hot spot at the surface of the particle acquire higher temperature upon their accommodations. Their desorption leads to the net momentum transfer to the particle. For deformed particles with defects and possible change in refractive index, the temperature gradient in microparticles caused by the illumination laser is rather random. This could result in “irregular” trajectories of a small fraction of particles, which are uncorrelated with the direction of the laser beam.

It can be easily shown that a temperature difference of a few degrees can already generate a considerable driving force. Assuming the particle radius is a and the effective

radius of a “hot spot” on its surface is a_* , the net momentum transferred to the particle can be written as $F \sim \pi a_*^2 n_0 T_0 (n_* T_* / n_0 T_0 - 1)$, where n_0 and T_0 are the number density and temperature of gas respectively, and T_* is the temperature of the “hot spot”. The density of absorbing atoms/molecules n_* is determined from the flux conservation $n_* \sqrt{T_*} = n_0 \sqrt{T_0}$ and thus $n_* T_* / n_0 T_0 = \sqrt{T_* / T_0} \geq 1$. From the balance with the Epstein drag $F_{ep} = 2\sqrt{2\pi} a^2 n_0 T_0 (u / v_{Tn0})$, where u is the particle velocity, we get $u / v_{Tn0} \sim (a_* / a)^2 (\sqrt{T_* / T_0} - 1)$. For $\delta T = T_* - T_0 \ll T_0$, we have $u / v_{Tn0} \sim 0.3 (a_* / a)^2 (\delta T / T_0)$. For a small temperature difference $\delta T / T_* \sim 10^{-2}$ at a small “hot spot” $(a_* / a)^2 \sim 0.3$, the particle velocity can already reach $u \sim 10^{-3} v_{Tn0} \approx 30$ cm/s.

For the particles with defects on its surface, as we see in figure 5, the defects may strongly enhance the light absorption and therefore generate such “hot spots”. Since the defects have various forms and distribute randomly on the surface of those deformed particles, the photophoretic force is not necessarily aligned with the direction of laser beam, resulting in the uncorrelated “irregular” motion of a small fraction of the particles, as shown in figure 4.

5. Conclusion

In conclusion, we presented the experimental observations of microparticles travelling in the particle cloud with “irregular” trajectories in PK-3 Plus laboratory on board the International Space Station. In those events, the particles were “trapped” in either the static or the scanning laser sheet under microgravity conditions. The irregular trajectories may have resulted from a combined effect of radiation pressure and photophoretic force exerted by the illumination laser. In the longitudinal direction, most of the “abnormal” particles move along the laser direction, driven mainly by the radiation pressure force. In the transverse direction, their motion is rather random, which may be caused by photophoretic force. To study the effect of photophoretic force quantitatively, we performed a series of dedicated experiments with a 2D complex plasma in the ground laboratory. The results show that the distribution of particle velocity has a larger tail at the high speed end with higher laser power, with the average kinetic energy of particles showing positive correlation with laser power. Comparing the velocity in the longitudinal and transverse directions with respect to the laser beam, little correlation was observed. This suggests that the photophoretic force indeed provides a major contribution to the drive of those fast-moving particles with “irregular” trajectories. The optical microscope observation suggests that the “abnormal” particles could be those with deformations or defects on their surface, which agrees with the theoretical estimation.

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