Observation of particle pairing in a two-dimensional plasma crystal

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The observation is presented of naturally occurring pairing of particles and their cooperative drift in a two-dimensional plasma crystal. A single layer of plastic microspheres was suspended in the plasma sheath of a capacitively coupled rf discharge in argon at a low pressure of 1 Pa. The particle dynamics were studied by combining the top-view and side-view imaging of the suspension. Cross analysis of the particle trajectories allowed us to identify naturally occurring metastable pairs of particles. The lifetime of pairs was long enough for their reliable identification.

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I. INTRODUCTION

A weakly ionized gas comprising dust or other fine solid particles is known as a complex, or dusty, plasma [1–3]. In experimental studies of complex plasmas the particle size is of a few nanometers to tens of microns. Immersed into a plasma, the particles charge up and interact with each other. It is a well-established fact that complex plasmas are able to self-organize, forming a highly ordered structure, plasma crystal, when the mutual interparticle interaction energy exceeds significantly their kinetic energy [4]. In the presence of gravity, a single-layer, or two-dimensional (2D) plasma crystal can form. Last two decades of studies showed that plasma crystals can be exploited as a useful tool to model or at least mimic at a kinetic level many phenomena as diverse as particle and energy transport in solids and liquids, crystal lattice plasticity, phase and structural transitions, etc. [1–2].

In plasma crystals, as in any other crystalline structures, point defects and dislocations are ubiquitous [5–6]. They may present an obstacle to performing some delicate experiments. Additionally, plasma crystals sometimes suffer from the presence of extra particles [7], which do not belong to the crystalline structure and can cause local instabilities and disturb the lattice. (Such particles are sometimes called “unstable”, “anomalous”, etc. or even, addressing their position in the flow of ions with respect to a particle layer, “upstream” or “downstream”, see, e.g., Ref. [8].) On the other hand, they can be successfully used, as the studies performed recently have shown, as an active agent in plasma crystal heating experiments [9, 10], as a convenient practical diagnostic tool allowing to test in the simplest way the complex plasma elasticity modules [11–13], or as a probe of the plasma electric field distribution [14].

The particles which constitute the main lattice of a crystal are called intralayer particles. This terminology is also used in the granular media [15] and colloidal [16] physics. The particles located between the layers in multi-layer crystals [9, 17] are naturally called the interlayer particles [16, 17].

The dynamics of interlayer particles are cardinaly different from those of the intralayer particles. For example, the dynamics of a single second-layer Delrin particle [15] free to move on top of a granular dimer lattice, or the cooperative permeation of string-like clusters in colloids of rods [16] reveal unusual features. In plasma crystal studies, the particles moving in a plane above a single-layer plasma crystal (they were termed upstream particles in Ref. [8]) reveal elements of “strange kinetics” [18] such as channeling and leapfrog motion [8].
A delicate repulsion-attraction balance can result in a strong correlation—pairing of the upstream particle with a neighboring intralayer one. To some extent, such kind of pairing resembles the famous Cooper electron pairs when the electron-phonon interactions produce a strong preference for singlet zero momentum electron pairs. Vertical pairing of two identical particles in the sheath of a rf discharge has been studied in Ref. [20].

In this paper, we report on the first direct observation of particle pairing and dragging occurring under natural conditions in a 2D complex plasma. Neither a torque, as in the “rotating wall” technique of Refs. [27, 29], nor a laser beam, as in the laser-dragging experiment of Ref. [21], nor any other method of external manipulation has been used. Using paired particles as a probe of the mutual interparticle interaction is one more possibility that is briefly discussed in this paper.

II. EXPERIMENTAL PROCEDURE

The experiments were performed in a modified version of the Gaseous Electronics Conference (GEC) rf reference cell [24] using argon at a pressure of 1 Pa and melamine-formaldehyde microspheres with a diameter of 9.19 ± 0.14 μm, a mass of \( m = 6.1 \times 10^{-13} \) kg, and a weight of \( mg = 6 \) pN, where \( g \) is the free-fall acceleration on earth. A stylized sketch of the experimental setup is shown in Fig. 1. A weakly ionized plasma is generated by applying a forward rf power of 15 W at 13.56 MHz to the lower disk-shaped rf electrode (corresponding to the self-bias voltage \( V_{dc} = -124 \) V). The microparticles, introduced into the plasma using a dispenser, formed a stable monolayer confined in the plasma sheath above the rf electrode. Optical ports and windows at the top and the side of the chamber provide access for the laser illumination and recording systems. Two digital cameras (a Photron FASTCAM 1024 PCI operating at 250 frames per second (fps) and a Basler Ace ACA640-100GM at 103.56 fps) recorded the microparticle positions and their dynamics and provided top- and side-view snapshot sequences subjected further to a standard particle tracking technique [31].

Side-view imaging is usually used in 2D plasma crystal experiments only as a complementary diagnostic. In the present study, we relied on it for our main results. Therefore, we first verified the side-view data, using the fluctuation spectra of the particle out-of-plane motion, see Fig. 2 as a cross-check. Additionally, the side-view camera was used to verify that our experiments were carried out with a (dominantly) single layer of particles.

III. COMPLEX PLASMA PARAMETERS

The plasma crystal parameters were evaluated using a well-developed method based on the particle tracking technique [31]. The lattice constant \( a \) was obtained from the first peak of the pair correlation function \( g(r) \). The neutral gas damping rate was estimated to be \( \gamma_E \approx 1.2 \) s\(^{-1} \) [34]. The small value of \( \gamma_E \) (compared to the characteristic frequency of the plasma crystal) assures weak frictional coupling of the particle dynamics to the ambient gas. Therefore, the particle motion is not overdamped and studying of the naturally occurring waves (fluctuations) can give reliable information about the lattice layer. The values of the particle charge \( Q \), interaction range \( \kappa = a/\lambda_D \) (where \( \lambda_D \) is the screening length), and the vertical confinement parameter \( f_v \) were estimated from the fluctuation spectra of particle velocity. These parameters are collected in Table I along with the parameter set adopted for numerical calculations performed for comparison reasons.

The experimental fluctuation spectra of the crystal obtained from either the top-view (TV) or side-view (SV) recording systems are shown in Fig. 2. Although both methods are widely used in complex plasma experimental studies (see, e.g., [13, 37] and the references therein), a cross-checking diagnostic has never been done before and the results of the TV and SV observations were never systematically compared. Below are important points of comparison that are worth to comment on: (i) the TV- and SV-spectra agree remarkably well with each other; (ii) the SV-spectra show systematically lower resolution in the wave numbers due to a significantly poorer spatial
TABLE I: Plasma crystal parameters measured from the top- and side-view recording data as well as the parameter set adopted for numerical calculations.

| Parameter               | Top view | Side view | Theory |
|-------------------------|----------|-----------|--------|
| Lattice constant, $a$ [μm] | $520 \pm 30^\alpha$ | $530 \pm 40^\beta$ | $500$  |
| Interaction range, $\kappa$ | $1.06$ | $1.06$ | $1.06$ |
| Particle charge, $Q$ [$10^3$e] | $15.0 \pm 2.3^\gamma$ | $15.3$ | $15.3$ |
| Vertical confinement   | $26 \pm 3$ | $24 \pm 3$ | $25$  |
| Parameter, $f_c$ [Hz]   | $31.0 \pm 2.2$ | $32 \pm 4$ | $31$  |
| Longitudinal phonon speed $v^a$, [mm/s] | $6.5 \pm 1.2$ | $6.5$ | $6.5$ |
| Transverse phonon speed $v^a$, [mm/s] | $26.43$ s, $t=0.19$ s | $31$ | $31$ |
| $\alpha$ in the central part of the crystal, measuring $14.6 \times 14.6$ mm$^2$; $\beta$ obtained from a row of particles that was well-aligned with the laser (left half of the top panel in Fig. 3); $\gamma$ for the intralayer particles, assuming no decharging of particles by ion wakes; $^a$ in-plane modes.

In our experiments, as in Ref. [8], a few upstream particles were wandering quasi-freely on top of the lattice layer along the channels made by the rows of ordered intralayer particles. From time to time, encountering a lattice imperfection blocking the channel, they strongly scattered and were forced to change the track direction, then moved again quasi-freely along another newly discovered path, and so on, covering a large area of the crystal. Usually, this process took quite a long time.

When an upstream particle happened to move in the vertical laser sheet, its trace was recorded by the side-view camera, as shown in Fig. 3. The travel path of an upstream particle is, on average, at the height of $\langle \Delta h \rangle \approx 0.2$ mm $\approx \frac{1}{10}E$ above the lattice layer (same as estimated in Ref. [8] using a top-view survey). In all cases shown in Fig. 3, the interaction scenario appears to be quite universal, passing normally through the following well-distinguished phases: initiation, repulsion, binding, and dragging. When an upstream particle comes too close to the channel wall or encounters a point defect, a strong interlayer collision between the top particle and a nearby intralayer one occurs. The bottom particle drops a little, allowing the “intruder” to pass over it. Then the repulsion is apparently replaced by attraction. The bottom particle starts to behave as if it was seized by the intruder, tending to be dragged with it. Since both particles are negatively charged, this is puzzling to some extent. The newly formed pair continues drifting for a while until the next strong collision would break it up.

V. COUPLED PAIRS AS QUASI-PARTICLES

The association of two particles in a pair strongly affects the motion of both particles: They start to accelerate as if the momentum was not conserved during their collision, see Fig. 4. This kind of action-counteraction imbalance is not surprising at all keeping in mind the following. First, the binding and subsequent dragging of an intralayer particle, the follower, actually is a direct manifestation of the ion focus (localized positive spatial charge or the ion wake) formed beneath the top particle,
which is in the upstream position in the pair. A negatively charged bottom particle is attracted by the ion focus while it is repelled by the negatively charged top particle. At the same time, the bottom particle continues to repel the top one whence accelerating it. The forces working to produce this motion are the plasma forces. Newly formed pairs behave as quasi-particles, which are (roughly) double-charged compared to the individual particles in the monolayer. This helps them to permeate through the lattice and to find an optimal path, e.g., inside a channel formed by the lattice particles, as the example shown in Fig. 4(right panel) demonstrates.

FIG. 4: (Color) Pairing of an upstream particle with intralayer particles. The left panel shows a space-time diagram assembled from 20 consecutive side-view images (each approximately 4.3 × 0.8 mm² in size). Time advances from top to bottom (in the range of 16.38 – 16.57 s, see Fig. 3), the time step is 0.009646 s. The cyan and red circles indicate the positions of respectively upstream and intralayer particles. The arrows indicate pairing events. The track of a long-living pair of particles is highlighted by two parallel dashed lines. The inset shows the dragging geometry. The right panel shows the top view of a different dragging event (assembled from 15 blended images; here, the illuminating laser sheet was shifted upward, which allowed to simultaneously record the upstream and intralayer particles). The filled circles indicate the positions of the upstream (cyan) and intralayer (red) particles in the beginning of a pairing event. The open circles indicate the particle positions 0.06 s later. The arrows indicate the resultant directions of particle motion.

Upstream particles move non-uniformly along their trajectories. For instance, in Fig. 3 (left panel) the velocity of such a particle is about 7 mm/s in the beginning and in the end of the travel path. However, it is more than twice larger, ⟨V⟩ ≃ 18 mm/s, when the particle becomes coupled, forming a close pair. This acceleration is due to the horizontal projection of the repulsion force between the coupled particles that is not completely canceled out. The average distance between the particles in the pair is r ≃ 0.36 mm, its horizontal projection (dragging distance) is δ ≃ 0.19 mm. On average, the dragged particle in the pair is kept at the height (Δh)drag ≃ 40 μm below the monolayer equilibrium position, experiencing therefore an extra force of external confinement. This gives a useful estimate of the z-component of the inter-pair repulsion force pressing it down: ⟨Fz⟩/mg = (Δh)drag/L_E ≃ 10%.

Given the approximately constant velocity of the pair, it is straightforward to roughly estimate the x-component of the dragging force: ⟨Fx⟩/mg ≃ 2(E_f /g ≃ 0.4%. It is about 25 times weaker compared to the vertical z-component, in good agreement with that measured in Ref. 8. Following Refs. 27, 42, the coupling between the particles in a pair can be conveniently interpreted through Hooke’s spring constant. Introduced by the relationship ⟨Fz⟩ = kδ, where δ is the dragging distance, it is k ≃ 900 eV/mm², noticeably well in line with that reported in Refs. 27, 42.

It is also worth noting that the lifetime of a pair is short, e.g., about 0.06 s for the pairs shown in Fig. 4. Their formation time (as well as the decomposition time) is even shorter, about 0.01 – 0.02 s. Therefore, these processes are controlled by much stronger coupling forces producing accelerations of the order of 50 – 100 cm/s², according to our estimates.

VI. CONCLUSION

We have observed for the first time the spontaneously forming mobile pairs of coupled particles in a 2D plasma crystal. This phenomenon is different from previously reported channeling, or “classical tunneling”. This observation was made possible by combined top- and side-view imaging of the dust particle suspension. We argue that the apparent self-acceleration of a particle pair is a direct consequence of the plasma wake effect. These naturally-occurring mobile pairs are metastable. They are, however, long-living enough for their reliable detection under our experimental conditions. The pairs we reported on in the present paper were formed by particles located initially at different heights. This helped to initialize the pairing process, because the mutual wake-mediated interaction was easily activated in this case. It is not strictly necessary for the particles to be initially at different heights, though. The pairing of particles is also possible, for instance, in the experimental situations when their vertical displacement becomes relatively large, thus enhancing the mutual wake-mediated interaction. Particle pairing is of primary significance in experimental studies of the later stages of the wake-mediated melting, as our preliminary observations have demonstrated.

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