Transition from Attractive to Repulsive Forces between Dust Molecules in a Plasma Sheath

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The electrostatic interaction of a system of two single dust particles in a plasma-sheath environment with flowing ions has been investigated quantitatively. It is shown that attractive net "binding" forces between the negatively charged particles exist, leading to the formation of a dust molecule. By laser manipulation of the dust particles, it is demonstrated that the attraction is asymmetric in such a way that it acts only on one of the particles. Moreover, the net forces between the particles can be reversibly changed between attraction and repulsion.

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The emergence of attractive forces between otherwise repelling particles by polarization of the surrounding medium is a well-known phenomenon in physics, e.g., for Cooper pairing or for molecular binding. Plasmas containing macroscopic "dust" particles have proven to be ideal model systems for the study of condensed matter systems on an "atomic" level [1]. In this Letter, the formation of dust molecules is presented and the nature of the interparticle forces are analyzed.

The dust model crystals are formed from monodisperse micrometer sized particles immersed in a plasma environment [2–5]. There, the particles generally acquire negative charges Q of the order of 10^3 to 10^4 elementary charges. Typically they are trapped in the sheath of radio-frequency (rf) discharges, where an inhomogeneous electric field $E(z)$ prevails that levitates the particles against gravity, where $QE(z) = mg$ (m being the particle mass and g the gravitational acceleration). Because of their mutual Coulomb repulsion, the particles can arrange in ordered structures, the plasma crystal. The properties of the plasma crystals have been widely studied in view of, e.g., the crystal structure [6–9], solid-liquid phase transitions [10,11], and wave propagation [12–14].

The observed unusual plasma crystal structure, where the particles of different layers are stacked on top of each other, was, on theoretical grounds, ascribed to attractive forces arising from the polarization of the plasma environment [15,16]. The ion streaming motion in the sheath causes an attractive wake behind the dust particles. With this reasoning, the dust particles behave like classical Cooper pairs [15]. From the same point of view, the polarization predicts a "binding" force for the formation of dust molecules [17].

From a more complete theoretical analysis of the nonequilibrium sheath environment, the interparticle forces were found to be both attractive and asymmetric in such a way that the attractive force is communicated only downstream along the ion flow [18–20]. The attraction is responsible for the vertical aligned crystal structure and the asymmetry drives the solid-liquid phase transition of the plasma crystal.

We report here on the experiments on the formation of a two-particle dust molecule. Repulsive and attractive particle interactions can be unambiguously derived by analyzing the reaction of one particle to the motion of the other. The two-particle system is also used as a quantitative force probe. A further advantage of this system is that many-body effects and collective modes as in plasma crystals are not present here. A selective external force that acts only on one of the two particles can be exerted with a focused laser beam, a technique which we applied recently for the excitation of waves in plasma crystals [13,14] and particle manipulation [21].

The experiments are performed in a parallel plate radio-frequency discharge in helium described in detail elsewhere [14] and schematically depicted in Fig. 1. The lower electrode is powered at 13.56 MHz with 5 W, and the upper electrode is grounded. The particles injected into the plasma are illuminated by an expanded laser beam. The scattered light is recorded by a video camera from the side (the camera is not shown in the figure). A second laser diode (690 nm, 40 mW) is used to manipulate the particles by the radiation pressure of

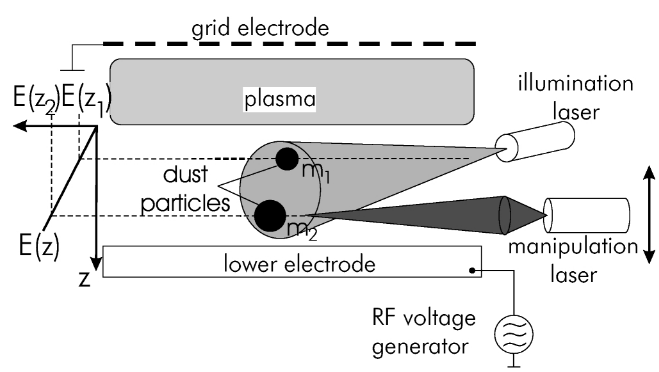

FIG. 1. Scheme of particle trapping in the plasma sheath of an rf discharge. Two particles of different mass find their equilibrium position at different heights above the electrode. Each of the particles can be manipulated by a focused laser beam that allows one to exert a controllable force onto either the upper or the lower particle.

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the focused laser beam. With the focus diameter of $\approx 200\ \mu$m, the radiation pressure is estimated to be of the order of 10^{-15} N.

We use two particles of different mass. The first one has a diameter of $a_1 = 3.47\ \mu$m and a mass of $m_1 = 3.3 \times 10^{-14}$ kg, and the second one is a cluster of two such particles sticking together by adhesion, thus $m_2 = 6.6 \times 10^{-14}$ kg. From the standard resonance technique [4,7], the charges of the two particles are determined ($Q_{1,2} \approx 2.000e$). Because of the different charge-to-mass ratio, the particles attain different vertical equilibrium positions z_1 and z_2 as indicated in Fig. 1. The force balance fixes the individual vertical positions of the two particles, thus no vertical stress acts on the two-particle system. Both particles, however, are free to move in the horizontal plane and the horizontal interaction forces between the particles can be directly deduced from an analysis of their horizontal motion.

The behavior of the two particles for different gas pressures in the discharge is shown in Fig. 2. Starting at a pressure of 130 Pa, the two particles are found horizontally separated from each other (Fig. 2a) as expected for a repulsing force. They do not separate to infinite distances because of a very weak horizontal confinement due to a curved electrode. The vertical distance is $d = 750\ \mu$m. When the discharge pressure is reduced to 55 Pa (Figs. 2b and 2c), the two particles suddenly jump into a vertically aligned structure as in the plasma crystal experiments [6,7]. The video sequence of the transition from the separated into the aligned state is analyzed in Fig. 3. The separated state is found at $t = 0$ s. Then the lower particle jumps towards the upper particle into the aligned state, the upper particle's horizontal position being almost unchanged ($t = 3.75$ s). Thereafter, the two particles move as a dust molecule to their final position ($t = 9$ s). The existence of the aligned state and the fact that the two particles move as a bound state clearly demonstrates that attractive forces are present here.

After increasing the discharge pressure to 130 Pa again, the aligned state is still preserved (Fig. 2d). A further pressure increase to 175 Pa finally leads to the breaking of the alignment where the particles jump from the aligned into the horizontally separated state (Figs. 2e and 2f). One should note that, during the breakup of alignment, the vertical positions of the particles do not change, thus indicating that indeed the vertical positions are determined only by the vertical force balance. The analysis of the corresponding video sequence is shown in Fig. 4. The particles are aligned at the beginning of the sequence, although the lower particle already is slightly shifted to the left ($t = 0$ s). At $t = 1.25$ s, the two particles separate within a fraction of a second. The breakup of the dust molecule clearly indicates that the particles experience mutually repulsive forces, so that the separated state is energetically favored.

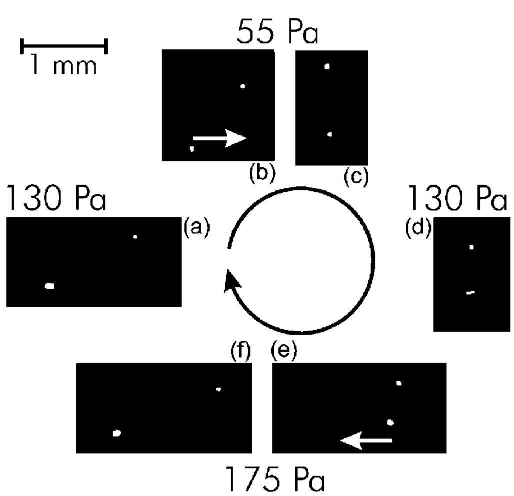

FIG. 2. Behavior of the two trapped particles with change of gas pressure in the discharge between 55 and 170 Pa.

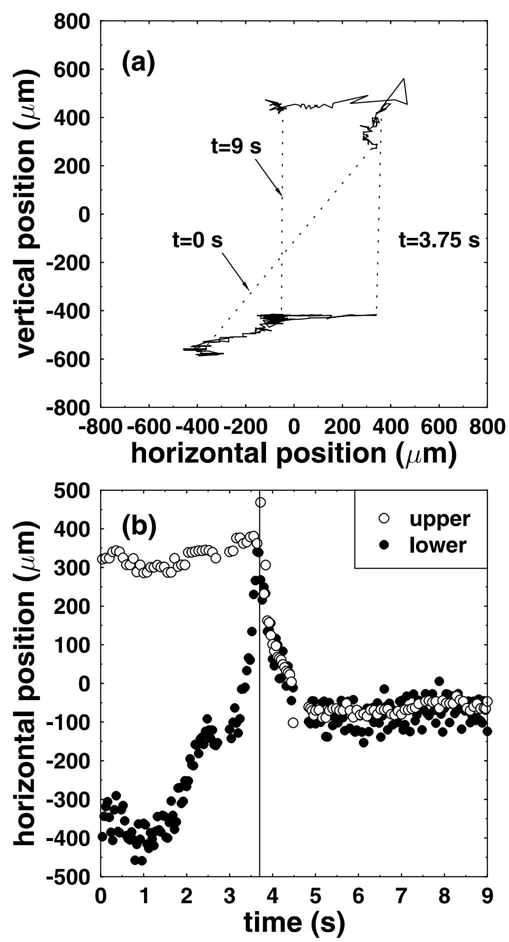

FIG. 3. Analysis of the video sequences during the jump into the aligned state, corresponding to Figs. 2b and 2c. In the upper figure, the particle trajectories as seen by the video camera from the side are shown. The dashed lines connect the particle positions at the given times. The lower figure shows the horizontal positions of the upper and lower particle with time.

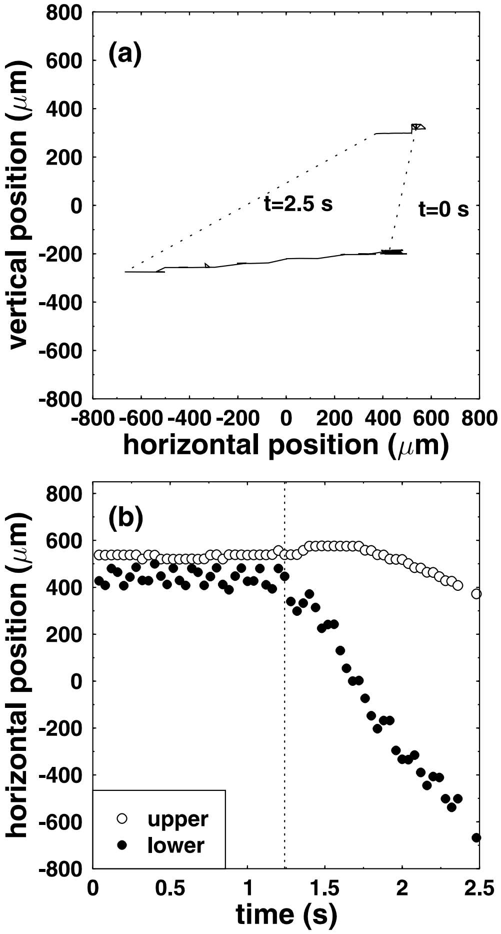

FIG. 4. Analysis of the video sequences during the breakup of the alignment, corresponding to Figs. 2e and 2f. In the upper figure, the particle trajectories as seen by the video camera from the side are shown. The dashed lines connect the particle positions at the given times. The lower figure shows the horizontal positions of the upper and lower particle with time.

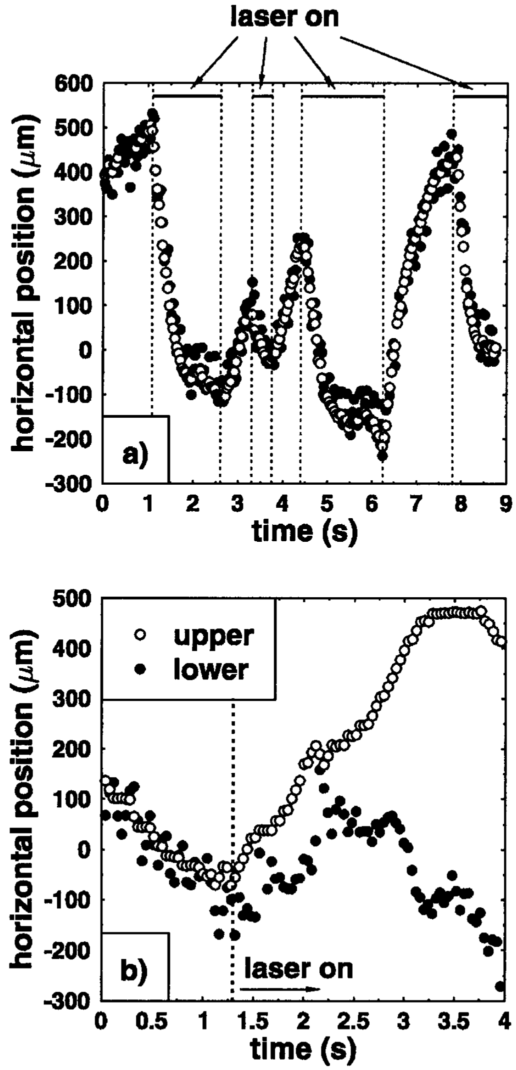

FIG. 5. Analysis of the video sequences in the aligned state at 55 Pa when (a) the upper and (b) the lower particle is pushed by the laser beam.

This loop (Figs. 2a–2f) between bound and separated states can be repeated perpetually. An attractive force between the two particles is found at low gas pressures, whereas at high pressures repulsive forces dominate in the system. In the intermediate pressure regime, one of the stable states (separated or aligned) is realized depending on the history of the system. Thus, here a hysteretic structural transition between these states is found.

In order to prove that the aligned state is due to attractive forces between the particles rather than being forced by any external confinement, the manipulation laser beam is focused either on the upper or the lower particle (see Fig. 1). The laser beam pushes one of the particles in a defined way and the reaction of the other particle is studied, from which the action of attractive forces becomes immediately evident.

First, the upper particle is pushed by the laser beam (see Fig. 5a). It is clearly seen that, when only the upper particle is hit by the laser, *both* particles react in the same way. The dust molecule stays aligned when being pushed around by the radiation pressure of the laser. The lower particle follows the upper particle, their horizontal positions being almost exactly the same. This behavior is a direct proof that the lower particle is subject to an attractive horizontal force mediated by the upper particle.

If, in contrast, the lower particle is pushed by the laser beam (Fig. 5b), the lower particle is shifted horizontally, but the upper particle does not follow the lower one. Moreover, the upper one is repelled from the lower one. Upper and lower particle are clearly separated horizontally. This definitely shows that the upper particle experiences no attractive force from the lower one. Hence, the interaction in the dust molecule is evidently attractive and asymmetric.

In order to obtain quantitative results from the experiment, the equation of motion of the two particles under the asymmetric interaction (see Fig. 6) is considered

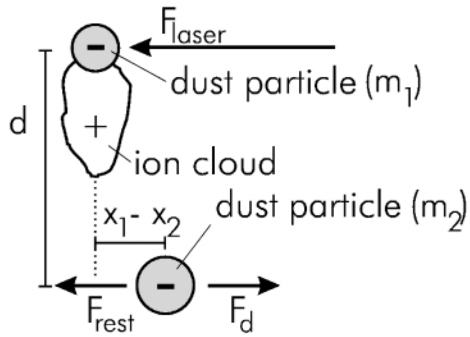

FIG. 6. Interparticle forces in the dust molecule.

now [20]:

$$
\begin{aligned}
m_1\ddot{x}_1 + m_1\beta_1\dot{x}_1 &= \frac{Q_1Q_2}{4\pi\epsilon_0 d^3}(x_1 - x_2)\,, \\
m_2\ddot{x}_2 + m_2\beta_2\dot{x}_2 &= (\epsilon - 1)\frac{Q_1Q_2}{4\pi\epsilon_0 d^3}(x_1 - x_2)\,,
\end{aligned}
\tag{1}
$$

where x denotes a small displacement from the vertically aligned position and β is the dust-neutral friction coefficient as determined from the resonance technique [4]. As shown from the experiment, the upper particle (index 1) experiences only Coulomb repulsion, whereas the lower (index 2) reacts to both repulsive and attractive forces. Here ϵ is the ratio of attractive to repulsive force.

A linear stability analysis of this system [18,19] immediately leads to the following inequality for the attraction parameter,

$$\epsilon > 1 + \frac{m_2}{m_1} = 3\,, \tag{2}$$

for the dust molecule to be stable. This means that the attraction provided by the ion cloud must exceed the repulsion between the dust particles by a factor of 3 for the experimental conditions here.

In the experiment, the strength of the attractive force in the dust molecule can be estimated from the force balance of the moving dust particles (see Fig. 6). The upper dust particle is set into motion by the radiation pressure of the laser beam (F_{laser}). Then the lower particle experiences a restoring force towards the aligned state provided by the attraction from the ion cloud. The restoring force F_{rest} is counterbalanced by the neutral drag force $F_d = m_2\beta_2\dot{x}_2$, which makes the lower particle lag behind the upper. Taking the velocity of the lower particle $\dot{x}_2$ during the first laser pulse (Fig. 5a at $t = 1$ s), the drag force, and thus the restoring force, is determined as $F_d = 1.6 \times 10^{-15}$ N. With the measured lag of $x_1 - x_2 = (40 \pm 20)\ \mu$m, the restoring force exceeds the Coulomb repulsion between upper and lower particles by a factor of $\epsilon - 1 = 20 \pm 10$. From this estimate, the dust molecule is found deep in the stable regime.

In conclusion, we have demonstrated the formation of dust molecules in an rf discharge due to net attractive and asymmetric forces between the dust particles. At high gas pressure, there is a transition from the bound state to the separated state. We attribute this transition, by comparing with the available plasma crystal models [19], to the reduced ion mean free path in the sheath. This leads to a destruction of the ion flow's polarization by randomizing ion collisions, which reduce the attractive force ratio below the threshold of $\epsilon = 3$.

Transferring these findings to plasma crystals, we have now verified that the attractive forces are responsible for the vertically aligned crystal structure. Furthermore, previous theoretical predictions [18,19] and experiments [9] on the force asymmetry in the plasma crystal are confirmed. From the experiments presented here, it is shown that this asymmetry is not a result of collective modes in plasma crystals, but is a fundamental property of the attraction by polarization in the supersonic ion flow. Forces are mediated by ion acoustic waves that can propagate only downstream along the flow.

Dust molecules therefore provide a new paradigm for attractive and repulsive forces arising from polarization of a surrounding flowing medium.

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