Vertical Pairing of Identical Particles Suspended in the Plasma Sheath

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It is shown experimentally that vertical pairing of two identical microspheres suspended in the sheath of a radio-frequency (rf) discharge at low gas pressures (a few Pa) appears at a well-defined instability threshold of the rf power. The transition is reversible, but with significant hysteresis on the second stage. A simple model which uses measured microsphere resonance frequencies and takes into account, in addition to the Coulomb interaction between negatively charged microspheres, their interaction with positive-ion-wake charges, seems to explain the instability threshold quite well.

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Recent interest in the properties of complex plasmas—charged monodisperse microparticles suspended in an electron-ion environment, is partially due to the possibility to model condensed matter phenomena on an "atomic" level [1–3]. The particles are charged negatively in a radio-frequency (rf) discharge (up to 10^3–10^4 elementary charges) and levitate in the lower plasma sheath, where gravity can be balanced by an inhomogeneous vertical electric field. The particles repel each other via a screened Coulomb interaction. Inside a confining horizontal potential they arrange themselves either into an ordered structure—the "plasma crystal," or form disordered, liquid- or gaslike, states [4–6].

At low gas pressures one has typically only a very few layers (as few as one) for these structures. Ion streaming motion in the plasma sheath produces a nonequilibrium environment, which is reflected in the properties of the complex plasma structures and makes them rather different from the known equilibrium ones [7,8]. One of the striking observations at sufficiently low gas densities (such that the ion-neutral mean free path is not small compared with a typical distance between microspheres) is an unusual "stacking" of the particles such that adjacent horizontal layers are located on top of each other [2,5,9]. This vertical "polarization" of the dusty plasma was ascribed to the attractive forces which result from the focusing of the ion flux under each particle (ion wake effect, see, e.g., Ref. [10]). Thus, the attractive interaction between the horizontal layers in a plasma crystal is asymmetric, such that the attractive force is communicated only downstream in the direction of the ion flow [10]. The same arguments were applied to predict a "binding" force for a possible "dust molecule" formation [11]. This theoretical prediction was verified recently by experimental studies of the competition between repulsive and attractive forces acting on two *different* dust particles levitated on *different* levels in the plasma sheath [12].

In this Letter we present experiments on a new instability which is observed for two *identical* microspheres suspended initially on the *same* level in the plasma sheath. In this Letter we report on the quantitative investigations involving (for simplicity) only two microspheres, which were undertaken to clarify the basic behavior for many identical particles. The instability appears first as a continuous (forward) bifurcation to a state of the vertically separated microspheres and then to a discontinuous vertical pairing. Depending on gas pressure, the last stage—the pairing—can be either strongly hysteretic (for lower pressure) or weakly hysteretic (for higher pressure).

The experiments were performed in a standard Gaseous Electronics Conference (GEC) rf reference cell [13] with the lower electrode powered at 13.56 MHz and the upper electrode grounded (see Fig. 1). Argon gas at various pressures between 1 and 7 Pa was used, and the discharge power (or a rf peak-to-peak voltage, $U_{\rm pp}$) was the control parameter of the instability described. The electron temperature and density were measured at the center of the discharge with a rf-compensated passive Langmuir probe. At these conditions the electron temperature T_e was found to be within 2–5 eV, while the electron density n_e ranged from 10^7 to 8×10^8 cm^{-3}, so that the electron Debye length varied from $\lambda_{De} \simeq 0.5$ mm (high $U_{\rm pp}$) to $\lambda_{De} \simeq 5$ mm (low $U_{\rm pp}$). These values of the Debye length are comparable to or larger than the separation distances measured, and thus Debye shielding plays only a minor role in this rather low-density plasma. The microspheres, suspended in the plasma, were illuminated by a laser sheet of about 100 μm thickness, and their imaging was performed by external CCD cameras from the top and from the side (in both cases via 300 mm

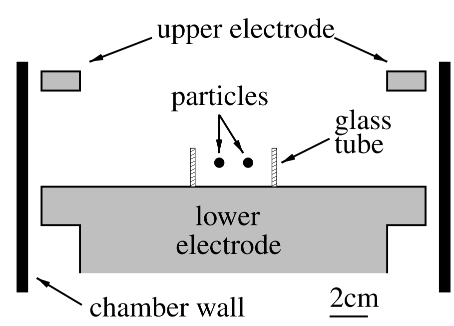

FIG. 1. Experimental setup using a quartz glass cylinder as a confining electrode.

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objectives). We used polystyrene (density 1.05 g/cm^3) microspheres with diameter 7.6 ± 0.1 μm and mass $M \simeq 2.4 \times 10^{-10}$ g. In order to confine the microspheres horizontally we placed a quartz glass cylinder of 20 mm height and 50 mm diameter on the lower electrode (see Fig. 1). In contrast to a copper ring normally used to confine particles, the glass cylinder produces extremely flat, almost "square well" confining potential. The latter conclusion is based on our observation of an unusually flat shape of 2D plasma crystal, particularly up to its edges, contrary to the trap formed by a standard metal ring. The square well confining potential provides us the opportunity to study two identical microspheres suspended in the plasma sheath on the same initial height.

In order to quantitatively investigate the pairing instability and to verify that it is indeed symmetry breaking, we conducted experiments with two identical microspheres at different pressures. Figure 2 shows how, at a pressure of 2 Pa, the relative position of one microsphere, which eventually moves below and then under the other (i.e., downstream) with respect to the eventual upstream microsphere, varies as the power (or U_{pp}) decreases. As long as U_{pp} exceeds the threshold value $U_{pp}^{th} \simeq 55$ V, both particles are located on the same level (initial position 1). When U_{pp} decreases below the threshold, vertical separation starts (position 2) and grows continuously up to position 3. A further small U_{pp} decrease causes the lower particle to "jump" to a position directly beneath the upper one and to create a vertical pair (final position 4). By reversing the process, i.e., increasing U_{pp}, the particles revert back to their initial horizontal configuration. The transition is strongly hysteretic with respect to U_{pp}. Further measurements showed that the threshold, U_{pp}^{th}, rapidly increases with pressure, while the relative width of the hysteresis decreases with pressure.

Figure 3 represents the forward (positions 1–5) and the reverse (positions 6 and 7) transitions at $p = 7$ Pa. The instability (vertical separation) starts at $U_{pp}^{th} \simeq 200$ V and develops continuously until position 4. Then a small U_{pp} decrease is accompanied by the discontinuous vertical pairing (position 5). When the voltage is increased back, the particles remain vertically paired until position 6 is reached, and then the lower particle jumps to position 7. In Fig. 4 the vertical separation distance is plotted against the control parameter, $U_{pp}^{th} - U_{pp}$, at $p = 7$ Pa. As in the low pressure case, the transition is divided into two stages: (i) continuous transition from the horizontal configuration and (ii) discontinuous, hysteretic transition to the final vertical pairing. It is worth noting that at the onset of the continuous transition strong *symmetrical* fluctuations of the vertical particle separation are observed in the experiments. This means that the pairing instability may be initiated with equal probability by either microsphere in the pair. Therefore, this transition is related to the symmetry breaking.

In the experiments both U_{pp} (at the fixed p) and p (at the fixed U_{pp}) were used as the control parameters of the instability. But in a subsequent analysis, presented below, it is much more convenient to use the resonance frequencies of vertical, ω_z, and horizontal, ω_r, oscillations of a single particle in the sheath as the control parameters. The resonance frequencies are certain functions of U_{pp} and p and can be determined experimentally [14].

We first examine the simplest model without wake effects. The model uses the measured values of ω_z and ω_r as the control parameters and provides an order of magnitude estimate for the instability threshold. We consider a pair of two identical particles of mass M and (negative) charge $-Q$, electrostatically confined in the harmonic potential

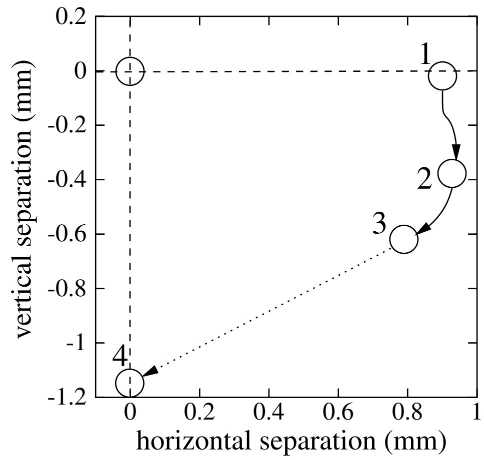

FIG. 2. Sequence of the relative particle positions during the pairing instability as U_{pp} decreases (pressure $p = 2$ Pa). The steps are (1) $U_{pp} \geq U_{pp}^{th} \simeq 55$ V—horizontal configuration, (2) $U_{pp} \simeq 45$ V, (3) $U_{pp} \simeq 37$ V—continuous separation, and (4) $U_{pp} \simeq 36$ V—discontinuous vertical pairing.

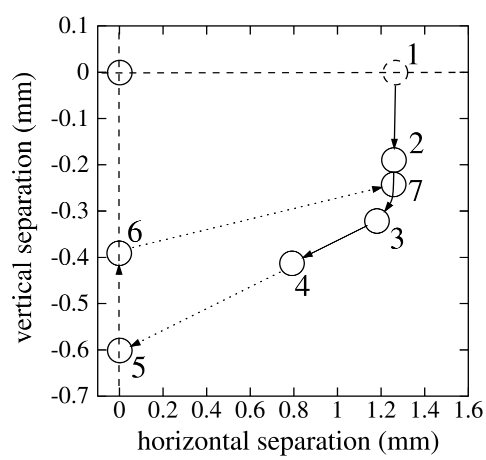

FIG. 3. Sequence of the relative particle positions during the pairing instability as U_{pp} decreases (1–5) and increases back (6 and 7) (pressure $p = 7$ Pa). For decreasing voltage the steps are (1) $U_{pp} \geq U_{pp}^{th} \simeq 200$ V—horizontal configuration, (2) $U_{pp} \simeq 110$ V, (3) $U_{pp} \simeq 85$ V, (4) $U_{pp} \simeq 78$ V—continuous separation, and (5) $U_{pp} \simeq 76$ V—discontinuous vertical pairing. For increasing voltage the steps are (6) $U_{pp} \simeq 93$ V—continuous vertical movement and (7) $U_{pp} \simeq 95$ V—discontinuous reversibility of the pair configuration.

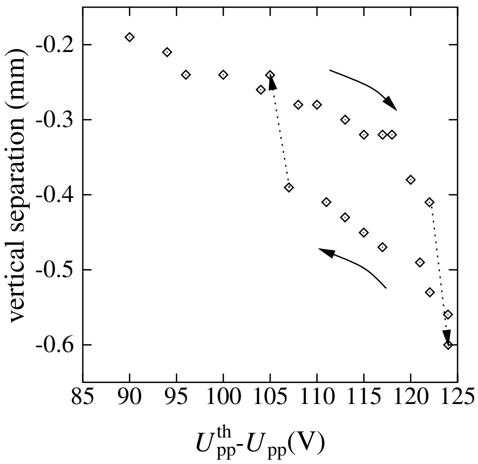

FIG. 4. Vertical separation of a particle pair vs the control parameter, $U_{pp}^{th} - U_{pp}$, for two stages of the pairing transition at $p = 7$ Pa. The hysteretic nature of the second stage is obvious from the plot. The arrows denote the direction of the variation observed.

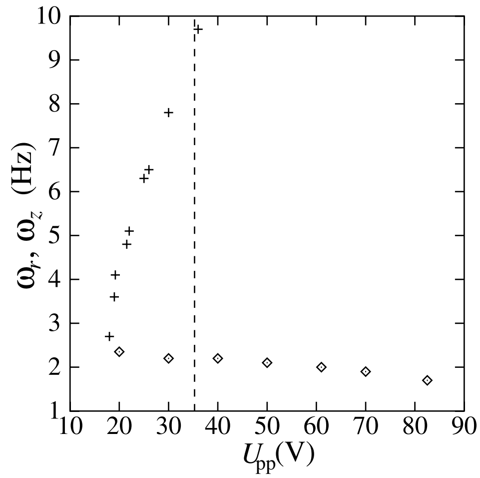

FIG. 5. Horizontal, ω_r (◇), and vertical, ω_z (+), resonance frequencies of a single particle as a function of U_{pp} (at a pressure $p = 1$ Pa). The vertical dashed line shows the threshold voltage $U_{pp}^{th} \simeq 35$ V (onset of the vertical separation).

well. If the particles are separated horizontally by R and vertically by δ (i.e., each particle is displaced at $R/2$ and $\delta/2$ from the center), then their energy in the confinement is $\frac{1}{4}M(\omega_z^2\delta^2 + \omega_r^2R^2)$. Since the Debye length is relatively large, the bare coupling energy of two unshielded particles, $Q^2(R^2 + \delta^2)^{-1/2}$, should be added to the confinement energy. This system has two stable configurations—horizontally (when $\omega_z > \omega_r$) or vertically (when $\omega_z < \omega_r$) aligned at distances of $R = (2Q^2/M\omega_r^2)^{1/3}$ or $\delta = (2Q^2/M\omega_z^2)^{1/3}$, respectively.

Our model is characterized in terms of the measured resonance frequencies. Thus, it is necessary to trace these frequencies as the control parameter, $U_{pp}^{th} - U_{pp}$, is varied. The typical dependence of ω_r and ω_z on the value of U_{pp} at $p = 1$ Pa is shown in Fig. 5. One can see that the frequencies converge rapidly as U_{pp} decreases. Comparison of Figs. 4 and 5 shows (albeit for different pressures) that the transition starts *continuously* when ω_z still *considerably exceeds* ω_r. This implies that the harmonic approximation with the isotropic particle field is too simple. Thus, we have to include the wake effect in its simplest form. For consistency in the nonlinearity analysis, we must also take into consideration the anharmonicity of the potential well in the vertical direction [15]. The corresponding expression for the total potential energy of a particle pair in a 2D confinement is

$$\mathcal{W}_{pair} \simeq \frac{M}{4}\left(\omega_r^2R^2 + \omega_z^2\delta^2 + \frac{\beta^*}{8}\delta^4\right) + \frac{Q^2}{\sqrt{R^2 + \delta^2}} - \frac{Qq}{2\sqrt{R^2 + (\Delta + \delta)^2}} - \frac{Qq}{2\sqrt{R^2 + (\Delta - \delta)^2}}. \quad (1)$$

In writing Eq. (1) the simplest two-microsphere model of the wake is used (see Fig. 6). We treat the excessive positive charge of the wake, q, as pointlike, located under the particle at distance Δ. We assume also that since both the particle and the wake charges vary only slightly with variation in R and δ, so that Q and q are approximately constant. The last two terms in Eq. (1) represent the particle-wake interaction, and cross terms of the particle-wake and wake-wake (qq) interactions are neglected [12]. Since the coefficients multiplying the terms δ^3 cancel out for a pair of identical particles, the lowest order vertical anharmonicity in Eq. (1) is the fourth order term with coefficient $\beta^* > 0$ but whose value is here unknown, since it was not feasible to measure it by technique used elsewhere [15].

The dependence $R(\delta)$ is determined from the equilibrium condition in the radial direction, $\partial\mathcal{W}_{pair}/\partial R = 0$:

$$\frac{2Q^2}{(R^2 + \delta^2)^{3/2}} - Qq\left(\frac{1}{[R^2 + (\Delta + \delta)^2]^{3/2}} + \frac{1}{[R^2 + (\Delta - \delta)^2]^{3/2}}\right) = M\omega_r^2. \quad (2)$$

The equilibrium value of δ is given by the condition $\partial\mathcal{W}_{pair}/\partial\delta = 0$. A linear combination of this condition and Eq. (2) gives

$$M(\omega_z^2 - \omega_r^2)\delta + \frac{M\beta^*}{4}\delta^3 + Qq\Delta\left(\frac{1}{[R^2 + (\Delta + \delta)^2]^{3/2}} - \frac{1}{[R^2 + (\Delta - \delta)^2]^{3/2}}\right) = 0. \quad (3)$$

First, we expand Eq. (3) to a third power in δ. Then we obtain from Eq. (2) an expression for $R(\delta)$ up to a second order expansion in δ (an even expansion) and substitute it into the coefficients of terms of first and third powers in δ in the expansion of Eq. (3). This approach provides the following expression which is a typical stationary equation for the vertical displacement as the order parameter:

$$\left(\frac{\omega_z^2}{\omega_r^2} - \Omega^2\right)\tilde{\delta} + \frac{\beta^* R_0^2}{4\omega_r^2}\tilde{\delta}^3 - \frac{35(\Omega^2 - 1)\tilde{\Delta}^2}{6(1 + \tilde{\Delta}^2)^2}\left(\frac{1 + \frac{8}{7}\frac{q/Q}{(1+\tilde{\Delta}^2)^{5/2}}}{1 - \frac{q/Q}{(1+\tilde{\Delta}^2)^{5/2}}}\right)\tilde{\delta}^3 = 0\,. \tag{4}$$

Here we have introduced the dimensionless parameters, $\tilde{\delta} = \delta/R_0$ and $\tilde{\Delta} = \Delta/R_0$ [normalized by $R_0 = R|_{\delta=0}$ from Eq. (2)], and the critical frequency ratio,

$$\Omega \equiv \sqrt{1 + \frac{6Qq\tilde{\Delta}^2}{M\omega_r^2 R_0^3(1 + \tilde{\Delta}^2)^{5/2}}}\,. \tag{5}$$

The instability criterion $(\omega_z/\omega_r)_{\rm cr} = \Omega$ is obtained from Eq. (4) by setting the coefficient of $\tilde{\delta}$ equal to zero. At $\omega_z/\omega_r > \Omega$, the energy $\mathcal{W}_{\rm pair}(\delta)$ has a minimum at $\delta = 0$ and the equilibrium configuration is horizontal. In the opposite case, $\omega_z/\omega_r < \Omega$, the $\delta = 0$ state becomes unstable, and the particles start separating vertically. From Fig. 5 we note that the separation starts at $(\omega_z/\omega_r)_{\rm cr} \simeq 4$ (when $U_{\rm pp} \lesssim U_{\rm pp}^{\rm th}$). Assuming $Q \sim q$ and substituting Eq. (2) at $\delta = 0$ into Eq. (5), one gets Ω as a function of $\tilde{\Delta}$ only. Then at $\tilde{\Delta} \sim 1$ we obtain $\Omega \sim 3$. Thus, the threshold of the pairing instability is adequately described by the model. A saturation of the order parameter, $\tilde{\delta}$, above the instability threshold in the first continuous stage of the transition (see Fig. 4) could be reached if the sum of both coefficients of $\tilde{\delta}^3$ in Eq. (4) is positive. By using the expression $\Omega(\tilde{\Delta})$ and the value of the ratio $Q/q \sim 1$ we achieve this condition at $\beta^* R_0^2/\omega_r^2 \gtrsim 7$, or $\beta^*/(2\pi)^2 \gtrsim 20$ Hz^2/mm^2. Our recent experiments [15] indicate that at pressures below ~10 Pa the anharmonic coefficients become relatively large: For example, $\beta/(2\pi)^2 \simeq 20$ Hz^2/mm^2 at $p \sim 1$ Pa and $U_{\rm pp} \simeq 70$ V. At smaller $U_{\rm pp}$, the whole vertical structure of the sheath changes dramatically, because the Debye length increases by the order of magnitude. Hence, the anharmonic coefficients should also increase rapidly as $U_{\rm pp}$ decreases, and it is plausible that β^* will provide the instability saturation. Then, as follows from Eq. (4), $\tilde{\delta} \propto \sqrt{\Omega - \omega_z/\omega_r}$ close to the threshold. However, failing an accurate experimental determination of the shape of the sheath potential for the actual conditions of the experiment, further testing of the model via the experimental value of δ is not feasible here.

The modeling of the second (discontinuous) stage of the instability is a rather complicated problem: When the lower particle approaches the wake, its actual "shape" becomes crucial, and the approximation of the pointlike wake charge cannot be used. This very nonlinear second stage will almost certainly involve nonlinear wake dynamics, and these can probably be adequately treated only by three-dimensional numerical simulations. We point out that the effect of the wake is stronger at lower p and $U_{\rm pp}$, when λ_{De} is larger. At the same time, the ion mean free path should exceed λ_{De} (otherwise, the wake effect weakens due to the ion scattering on neutrals). Both these requirements are well satisfied in our experiments.

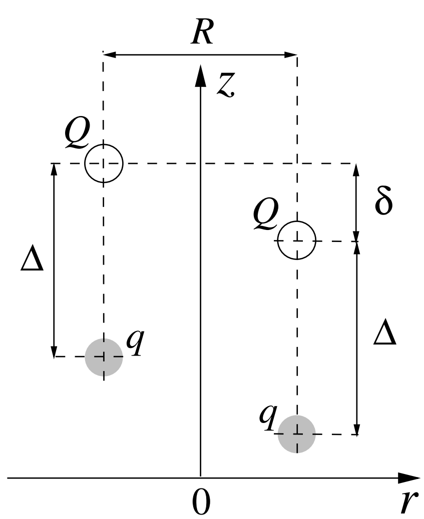

FIG. 6. Schematics of the simplified model of the wake potential.

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