Evolution of a Dust Void in a Radio-Frequency Plasma Sheath

R. P. Dahiya,[1,*] G. V. Paeva,[1] W. W. Stoffels,[1] E. Stoffels,[1] G. M. W. Kroesen,[1] K. Avinash,[2,†] and A. Bhattacharjee[2]
[1]*Department of Physics, University of Technology, 5600 MB Eindhoven, The Netherlands*
[2]*Department of Physics and Astronomy, University of Iowa, Iowa City, Iowa 52242*
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The onset and growth of a dust void are investigated in a radio-frequency (rf) sheath of a capacitively coupled argon plasma. A circularly symmetric void emerges and grows with increasing rf power and pressure in the central region of the dust cloud levitating in the sheath. Experimental measurements of the void diameter are compared with the predictions of a simple phenomenological theory, based on a balance of forces on dust grains.

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A complex plasma (also known as a colloidal or dusty plasma) consists of electrons, ions, and a dispersed phase of microparticles (called dust grains). Because of their large electric charge, dust grains are subject to mutual Coulomb interactions. Several laboratory experiments [1–4] have demonstrated the formation of organized crystalline structure in dust clouds trapped in the sheath of low-density plasmas. In a recent microgravity experiment [5], condensed states were produced with a liquidlike phase of dust adjacent to crystalline regions. A remarkable feature of these experiments is that often, under a wide variety of conditions, the dusty plasma forms a stable void, which is a local dust-free region with sharp boundaries.

Observations suggest that the tendency of a dusty plasma to form a void is fairly robust and independent of the initial process that triggers it. In laboratory experiments where the particles are grown *in situ* [6,7], formation of the void is preceded by a sudden onset of a filamentary mode [7]. In the microgravity experiment, on the other hand, where microparticles are already of sufficiently large size, formation of the void appears to occur without any initially unstable phase [5]. In all experiments where the void is seen [5–8], the void size is observed to increase with radio-frequency (rf) power.

Various mechanisms have recently been proposed to explain the dynamical process of void formation. These include the thermophoretic force due to temperature gradients in the microgravity systems [5], or the ion drag on dust particles in the laboratory [7,9,10]. A number of theoretical calculations have considered the role of the ionization instability [11–14] in the presence of ion drag and collisions of ions and dust with the background neutral gas as a possible dynamical trigger for void formation.

In this work, we systematically investigate the evolution and dynamics of voids in dust clouds suspended in a radio-frequency plasma sheath. Specifically, we study the variations of the void size by varying several external parameters, such as input rf power and argon gas pressure. Formation of the void is seen as a consequence of the compression of the dust particle cloud. This shrinkage can be triggered by a variety of physical interactions, be it mutual attraction between particles or external compression due to, e.g., ion wind. We consider a global phenomenological model based on the dynamical balance between the repulsive screened Coulomb (Yukawa) force and other cohesive forces that may be present between the grains. Using this model, the volume of the particle cloud and its behavior with varying external parameters can be determined, and the predictions appear to be in good agreement with experimental observations.

Experiments are performed in a capacitively coupled 13.56 MHz rf plasma reactor schematically shown in Fig. 1. An rf powered electrode with a centrally symmetric 3 cm diameter, 3 mm deep hole is horizontally mounted at the bottom of the vacuum vessel. The vessel is electrically grounded and acts as a counterelectrode for the rf power coupling. Melamine formaldehyde (MF) dust particles of 9.8 μm diameter are injected in an argon plasma using a horizontally tractable particle injector. The injected dust particles acquire negative charge and are trapped in the sheath. They are confined within the electrostatic potential well above the hole in the powered electrode. An argon ion laser beam sheet is used to illuminate the particle cloud, which is then visualized via the scattered laser light. Magnified video images of the particle cloud are recorded by means of a video camera viewing from the top of the system. Gas injection and pumping system are positioned in the same plane close to the chamber top. This prevents the gas flow driven neutral drag on the particle cloud. A mass flow controller is used for regulating the argon gas flow rate, which is kept 10 sccm (sccm denotes cubic centimeter per minute at STP) during the experiment. Evolution and development of the void is followed as a function of the rf power and working pressure.

The particle cloud levitating in the rf sheath region of an argon plasma forms a stable Coulomb crystalline structure at 0.13 mbar pressure and 3 W power. In the crystal, a centrally symmetric void evolves with the increase of gas pressure (p_g) and/or rf power ($P_{\rm rf}$). This void, which has sharp and well-defined boundaries, keeps expanding with increasing pressure and/or power. We do not observe the initially unstable phase preceding the void formation

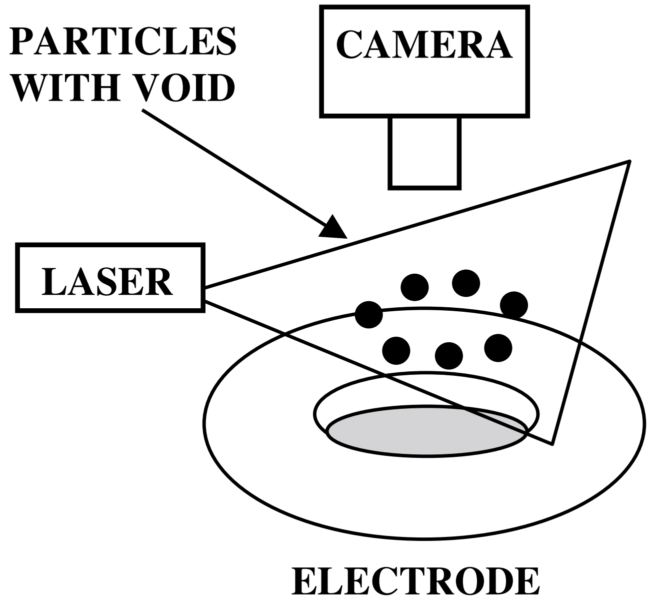

FIG. 1. Schematic diagram of the experimental system. The 10-cm-diameter rf powered electrode has a 3-cm-diameter and 3-mm-deep cylindrical hole. A void can evolve in the dust cloud trapped in the curved sheath above the hole. The particle cloud visualized by a horizontal sheet of Ar ion laser beam and images viewed from the top via scattered laser light are recorded using a video camera.

presumably because our particles are substantially larger than the ones formed *in situ* in the plasma [6,7]. The Coulomb crystal and the void, observed in our experiments, are shown in Figs. 2(a) and 2(b), respectively, for argon plasma at 0.13 mbar pressure at 3 and 10 W power, respectively. A similar void appears by increasing pressure at constant power.

We have evaluated the outer diameter of the dust cloud and the void diameter from the video images. These are shown in Fig. 3 as a function of pressure for 10 sccm flow of argon and 10 W rf power. The experimentally measured outer diameter of the dust cloud continuously increases from 4.5 mm at $p_g = 0.06$ mbar till it is 7.9 mm at $p_g = 0.1$ mbar, the pressure at which the void emerges. Initially, the void expands rapidly with pressure till $p_g = 0.13$ mbar, and then it grows slowly. At higher pressures the diameter of the void approaches the outer diameter of the whole dust cloud, which shows that the volume occupied by particles is reduced to a mere ring. At pressures above 0.16 mbar the particles are observed to align in rings in between void diameter and the outer diameter of the cloud. The cloud, having 18 mm outer diameter at the highest pressure applied in our experiments, was still far away from the edge of the radial potential trap, induced by the 30 mm deep hole in the powered electrode.

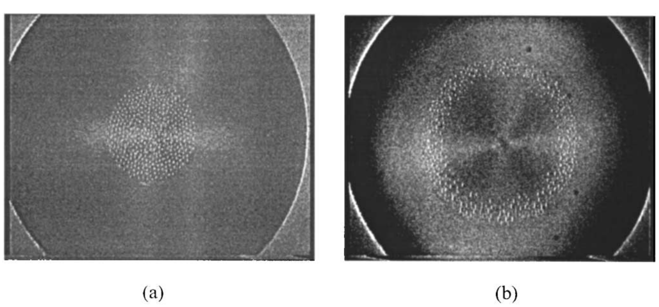

FIG. 2. (a) Crystalline structure of 9.8 μm diameter MF dust particles trapped in an argon rf sheath at 0.13 mbar and 3 W power, and (b) centrally symmetric 7-mm-diameter void formed in the dust cloud in an argon rf sheath at 0.13 mbar and 10 W power. Depression in the electrode (30 mm diameter, 3 mm deep) is visible as a bright circle on the periphery. The pictures are recorded by video camera viewing from the top of the experimental system in a plane normal to the page.

Figure 4 shows the outer cloud diameter and void diameter variation with rf power at 0.11 mbar pressure. Void formation occurs for powers above 15 W. At high powers, shrinkage of the particle cloud into a ringlike structure is observed. Shrinkage of the particle cloud can express itself in the formation of a dust-free region.

One can think of various mechanisms leading to dust cloud compression and attendant formation of the void. Thermophoretic forces have been suggested [5]. On the other hand, the presence of the radial potential trap in our configuration is a special feature, which can facilitate formation of a central void in the particle cloud. The depth in the electrode induces a curvature in the sheath in the radial direction, which in turn deflects positive ions advancing from the plasma glow towards the electrode. Thus, because of the local sheath geometry, positive ions acquire a radial velocity component, which gives rise to a radial ion

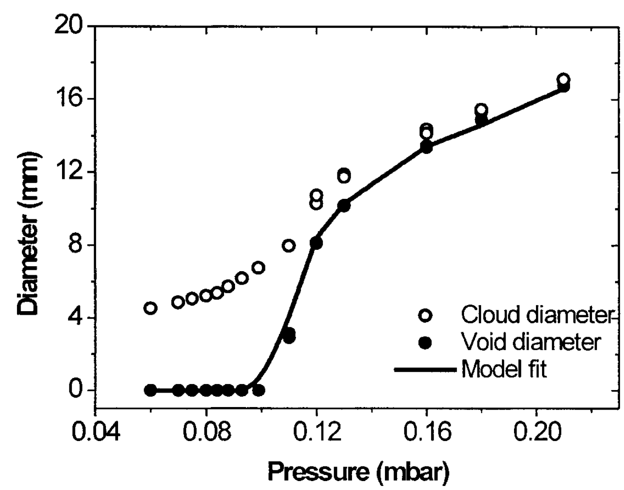

FIG. 3. Evolution of the dust void and radial expansion of the particle cloud in an argon rf plasma sheath as a function or argon pressure. Ar flow rate 10 sccm, rf power 10 W. The dust cloud consists of a few hundred particles. Solid line represents the void diameter based on the model.

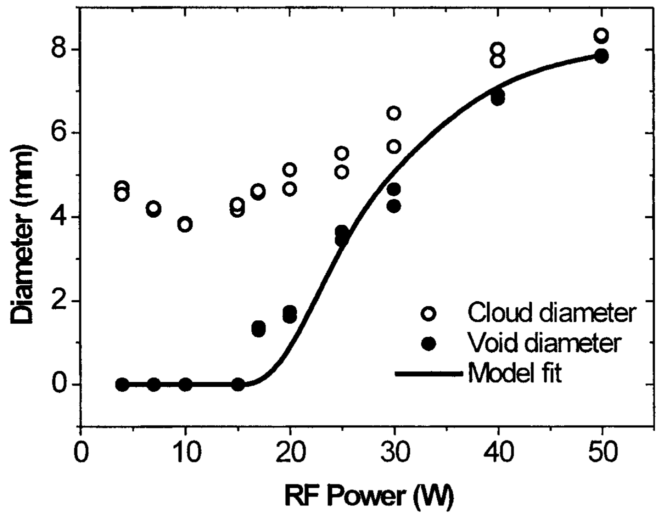

FIG. 4. Variation of the void diameter and cloud outer diameter as a function of rf power. Pressure is 0.11 mbar, and Ar flow rate is 10 sccm. The dust cloud consists of about 25 particles. Solid line represents the void diameter based on the model.

drag force pushing the particles towards the edges of the circular trap.

It must be stressed that the void is merely a form of appearance of dust cloud compression, which is governed by various physical processes. In our geometry the external compression by the ion drag force may play a role, but, on the other hand, the mutual interactions between the particles within the cloud can establish the optimal size of the dust cloud. Therefore, in order to obtain the volume occupied by particles, we propose a simple formalism, which takes into account the balance between the repulsive Yukawa force and cohesive forces.

The particular cohesive force we choose to work with is a van-der-Waals-like mean field that is derivable from well-known principles of statistical thermodynamics applied to strongly coupled Yukawa systems [15]. This cohesive force is attractive, as opposed to the repulsive Yukawa force between dust particles, and is due to the potential energy of the background plasma that, on average, neutralizes the charge of the dust particles. It is well known that this "mean-field" contribution has to be added to the pairwise potential energy calculated from molecular dynamics simulations of Yukawa particles in order for the simulation results to agree with the free energy calculations from first principles [16]. In the context of one-component plasmas, these contributions are also known as "bridge functions" [17]. Based on similar ideas, a detailed thermodynamic model for liquid, vapor phases, and void formation has been given recently [18]. It has also been shown that these contributions can drive reentrant phase transitions and phase coexistence in colloids [19]. We do not suggest that such a cohesive force is the only relevant one in the experiment, but rather use it as a proxy for other cohesive forces such as pairwise thermophoretic force, or shadow force [20], that might also be important but for which there are as yet no direct and conclusive measurements in the experiment. Thus, the proposed approach allows one to predict the trends in the behavior of the dust cloud without commitment to one particular type of interaction, and it can be easily generalized for any other dusty plasma system.

For dusty plasmas, the normalized potential energy (PE) per particle due to the cohesive field described above can be expressed as [16]

$$W_1 = -3\Gamma_A/2\kappa^3, \tag{1}$$

where $\Gamma_A = Q^2/4\pi\varepsilon_0\lambda_D T$, Q is the particle charge, T is the kinetic temperature, and $\kappa = a/\lambda_D$, λ_D being the Debye length. This is the potential energy required to assemble the smooth plasma background in the field of N grains [15]. The PE per particle due to the Yukawa field is given by

$$W_2 = k\Gamma_A \exp(-\kappa)/\kappa, \tag{2}$$

where k is the number of near neighbors. Since interacting grains try to attain a configuration of minimum PE, an energetically optimal interparticle distance will be established. This distance is given by $a = a_m \equiv \kappa_m\lambda_D$, where κ_m can be obtained by minimizing $W = W_1 + W_2$ with respect to κ. Interestingly, once the existence of κ_m is given, our arguments are not very sensitive to the exact functional form of the cohesive field. Using this model, the volume of a cloud consisting of N particles in its minimum PE state can easily be determined: $V_m = (4\pi/3)N(\kappa_m\lambda_D)^3$. This can be compared to the volume available to the particles: $V = \pi r_c^2 h$, where r_c is the cloud diameter and h is the cloud thickness. The latter is jointly determined by the rf power P_{rf}, the neutral gas pressure p_g, and, presumably, the external confining potential in the experiment. On the other hand, V_m varies inversely with p_g or P_{rf} because the plasma density (which enters the parameter λ_D) is found to scale approximately linearly with p_g or P_{rf} under our experimental conditions [21].

The measured dependence of the cloud diameter on p_g or P_{rf} is depicted in Figs. 3 and 4, respectively, where the cloud diameter is seen to increase (except an initial contraction with rf power) with these parameters. Now at low p_g or P_{rf}, we have $V_m > V$, and the state of minimum PE is inaccessible. In such a case, the grains, on an average, see a mutually repelling field in a volume V, and the only possible configuration is a homogeneous state containing a space-filling crystal or a so-called Barnal fluid [18]. As p_g or P_{rf} is increased, V and r_c increase (Figs. 3 and 4) and V_m decreases. For large p_g or P_{rf}, we have $V > V_m$, and a void of volume $V_\nu = V - V_m$ develops. Thus the condition $V = V_m$ defines a threshold for void formation. Using this model, the void diameter can be calculated for given N as a function of the cloud diameter, p_g and $P_{\rm rf}$.

Once V_m is normalized on one of the experimental points, the behavior of the particle cloud can be fairly accurately simulated. Note from Figs. 3 and 4 that the model predictions of the void diameter represented by the solid line and the measurements denoted by solid circles have a good match and also that the threshold for void formation as a function of pressure and power corresponds to the experimental findings. It should be noted that the model predicts only the volume of the particle cloud and not its shape in the state of minimum PE. Note that the theory is based on a force balance in a stationary state, so the energy minimization principle can be applied even though it is a thermodynamically open system. We emphasize that our assumption regarding the dominant nature of the cohesive field may be an approximate one, and that a more accurate model for the cohesive field, validated by experimental tests, may provide a better fit for the data.

In conclusion, we have investigated the evolution and growth of a dust void in an rf plasma sheath. Cohesive forces between dust particles compete with the repulsive Yukawa potential, which determines the optimal interparticle distance and, consequently, the volume occupied by the particles. Under many conditions this volume is smaller than the total available space determined by external constraints (e.g., plasma potential). This can lead to the formation of a void within the particle cloud. We observe that the volume of particle cloud decreases with increasing gas pressure and rf power, and a circular void is created. Measured void diameter is in good agreement with its theoretical values obtained on the basis of balance of forces between the dust grains.

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*Permanent address: Indian Institute of Technology, New Delhi.
†Permanent address: Institute for Plasma Research, Bhat, Gandhi Nagar.

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