Fast and interrupted expansion in cyclic void growth in dusty plasma

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Fast and interrupted expansion in cyclic void growth in dusty plasma

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Abstract

Low-pressure acetylene plasmas are able to spontaneously form dust particles. This will result in a dense cloud of solid particles that is levitated in the plasma. The formed particles can grow up to micrometers. We observed a spontaneous interruption in the expansion of the so-called dust void. A dust void is a macroscopic region in the plasma that is free of nanoparticles. The phenomenon is periodical and reproducible. We refer to the expansion interruption as ‘hiccup’. The expanding void is an environment in which a new cycle of dust particle formation can start. At a certain moment in time, this cycle reaches the (sudden) coagulation phase and as a result the void will temporarily shrink. To substantiate this reasoning, the electron density is determined non-intrusively using microwave cavity resonance spectroscopy. Moreover, video imaging of laser light scattering of the dust particles provides their spatial distribution. The emission intensity of a single argon transition is measured similarly. Our results support the aforementioned hypothesis for what happens during the void hiccup. The void dynamics preceding the hiccup are modeled using a simple analytical model for the two dominant forces (ion drag and electric) working on a nanoparticle in a plasma. The model results qualitatively reproduce the measurements.

Keywords: dusty plasma, dust void, microwave cavity resonance, nanoparticles, complex plasma, acetylene plasma

Online supplementary data available from stacks.iop.org/JPhysD/48/035204/mmedia

(Some figures may appear in colour only in the online journal)

1. Introduction

Dusty plasmas, i.e. plasmas containing solid nanoparticles, are ubiquitous. In processing plasmas, for example, dust particle formation is often an unwanted side effect [1, 2]. In addition, dusty plasmas impact several other research fields as well. Firstly, dusty plasmas are frequently encountered in space [3]. For instance, planetary rings and the tails of comets are dusty plasmas. Secondly, nanoparticles are encountered in fusion devices and for instance pose problems in terms of radioactivity and chemical reactivity, but they can also interfere with the discharge itself [4, 5]. Although exhibiting extremely high temperatures, a fusion plasma even allows for particles to grow inside such a device. Thirdly, nanoparticles in the low-pressure plasma environment of the projection optics and the wafer stage of (extreme ultraviolet) optical

lithography devices can severely limit the image quality and therefore also negatively impact overall process yield and life-time for optical devices [6, 7]. Dusty plasmas also proved useful to the study of fundamental physical phenomena, in particular phase transitions [8], where dusty plasmas exhibit the remarkable characteristic of mimicking nanophysics at the micro scale. More specifically, in dusty (or complex) plasmas, particles can arrange themselves in liquid- or crystal-like structures [9], which also may exhibit phase transitions. For particle sizes roughly on the order of the lights wavelength, the particles can easily be visualized using laser light scattering, enabling individual particle positions and trajectories to be resolved by camera. In this way, physics that for ordinary liquids and crystals takes place at the nanometer scale, can now be studied using relatively simple techniques at the micro scale.

In the laboratory, dusty plasmas can be created in two ways. Either by actively introducing premanufactured nanoparticles/microparticles in the plasma or by letting the plasma grow the particles itself by supplying a suitable chemical reactive gas. Several gasses have shown to initiate dust particle formation, such as hydrocarbons [10–12], fluorocarbons [13] and silane [14, 15].

Following the early experiments of Boufendi, Bouchoule, and others in the early 1990s [16–18], dust particle formation in reactive plasmas has been explained by a four-step process. First, the precursor molecules get dissociated into particles that polymerize into bigger clusters. The likely candidates to survive in a plasma environment are the anions which are actively trapped within the glow region, whereas the cations are attracted by the discharge walls and the neutrals are lost by diffusion. A brief period (several ms) of nucleation follows when the clusters reach a critical size (about 2 nm). At this stage, particles do not carry a high charge and can therefore also attain a positive net charge due to stochastic charging. In the third step these particles abruptly coagulate when they reach a critical density, pushing proto-particles together and ‘fusing’ them as long as they are not electrostatically repelling each other. The result is the formation of relatively big particles of tens of nanometers. Due to their size they start to accumulate a high net negative charge akin to the charging of any macroscopic surface in contact with plasma. Coagulation coincides with the so-called transition from an α (pristine) to γ' (dusty) phase. It involves a drop in electron density and a jump in electron temperature/energy [19–21]. In the fourth phase, dust particles continue to grow by surface growth, sometimes called accretion.

Recently, however, models of the group of Girshick [22, 23] and in particular [24], seem to suggest a slightly different mechanism. Instead of a brief burst of nucleation, the authors propose that nucleation continues in regions sufficiently free of nanoparticles. This locally produces small neutral nanoparticles. Coagulation is now of a very specific kind: ‘scavenging’ of small neutral particles by the larger negatively charged nanoparticles formed previously. This scavenging is greatly enhanced by the image potential a charged particle induces in a neutral particle.

A common observation in laboratory dusty plasmas is the appearance of a dust-free zone, generally referred to as the dust void. Usually, the dust void size and shape do not vary in time if dust particles of a fixed size are present in the discharge. An example of such an experiment can be found in [25]. However, dust voids show a dynamic behavior (such as expansion and contraction) if the suspended nanoparticles are growing in the plasma. The cyclic formation of dust particles in combination with the development of a void has been observed earlier in argon–silane [15], in argon–acetylene [11, 12, 26], and in argon–methane [11, 26] low-pressure plasmas, as well as in other types of radio-frequency discharges [27].

A dust void develops as a result of momentum transfer from flowing ions to dust particles, either by a direct hit or by a deflection of the ion trajectory. In both cases this results in a net force pointing in the direction of the ion flow. Ions, whenever and wherever they are created, will follow the ambipolar electric field. As a result, the force, formally called the ion drag force, points radially outwards with respect to the point of zero ambipolar electric field in the plasma. This point will generally be the

center of the dust void. The ion drag force is of great importance to dusty plasma physics. This force, and other drag forces, continue to be the focus of many (theoretical) studies [28–31]. In a recent paper [24], the authors were able to reproduce the formation of a dust void in their simulations and observed a greatly increased nucleation rate within this dust-free region.

Other forces working on dust particles could stem from gravity, neutral drag, thermophoresis, radiation pressure, and electric fields. The latter can be divided into two parts: resulting from the plasma potential (ambipolar field) and from the mutual (screened) Coulomb interaction between the dust particles. Interestingly, not every force scales the same with particle radius [32, 33]. As a result, the directionality of the resultant force can change abruptly when the particle radius exceeds a critical value. Unless an electrode is heated, thermophoresis is usually not important. Radiation pressure can be neglected if no powerful lasers are used. Neutral drag is insignificant if a closed vessel is employed. Some researchers eliminate also gravity by conducting experiments in micro-gravity (satellites [34, 35], parabolic flights [31]).

Recently, Couëdel *et al* [36] reported the observation of a self-excited void instability showing some similarities with the observations presented in this Paper. In that work, nanoparticles originated from sputtering of a polymer layer present on the electrodes in an otherwise pristine argon discharge. Many features of the void instability were not fully understood. In this work, nanoparticles are formed from the reactive gas acetylene supplied to our discharge. Most importantly, we employ microwave cavity resonance spectroscopy. This not only enables us to measure the electron density non-intrusively during the whole experiment, but we can selectively probe the region inside the void. We also explain the observed void dynamics by constructing an analytical model for the forces working on the nanoparticles. Combined, the model and powerful experimental techniques enable us to shed light upon the peculiar and interesting void dynamics.

In this Paper, we report the observation of a spontaneous interruption in the dust void expansion, which we refer to as expansion hiccup. We infer this is related to the coagulation of a new batch of dust particles that started growing in the dust-free region of the expanding void. The Paper is structured as follows: (1) a description of the experimental setup and methods, (2) an in-depth discussion of the experimental observations, (3) the development of an analytical force model describing the first two phases of the void expansion, (4) an interpretation of this model, as well as the remaining experimental observations, and (5) conclusions. The Paper also contains an appendix, summarizing the expressions for important forces working on a plasma-confined dust particle. It also describes the remaining details for the analytical model.

2. Experimental details

The experimental setup is schematically shown in figure 1. The plasma is generated in a cylindrical aluminum discharge chamber (cavity) with a volume of about $1.6 \times 10^{-3} \text{ m}^3$ (radius/height = 1.35), residing within a stainless steel vacuum vessel. A mixture of argon (17.4 sccm) and acetylene (2.3 sccm) enters

Figure 1. Schematic of the experimental setup. A dusty plasma is generated in a cylindrical discharge chamber/microwave cavity (1) residing in a vacuum vessel (2). Further details can be found in the text below.

the chamber through the cavity's top lid shower head. This lid also serves as the powered electrode and is capacitively coupled to an RF generator and matching circuit (L-type). A computer controlled butterfly valve keeps the pressure within the chamber constant at 10 Pa. To allow optical access to the chamber's volume, the chamber's side wall is made of a metal mesh (thread width 0.35 mm, mesh size 1.06 mm, translucency 57%). The bottom of the cavity is a disk containing a regular grid of 1 mm sized holes. In addition, a 2 mm wide slit along the diameter is used to guide a vertical laser sheet through the chamber, see below. This disk and the side walls are grounded and insulated from the powered top lid.

To study the dynamics of the low-pressure dusty argon-acetylene RF plasma, several temporally resolved and in addition spatially resolved measurement techniques are employed. The free-electron density is determined with microwave cavity resonance spectroscopy (MCRS) in a manner similar to [37]. In short, with this method one excites resonant modes of the electromagnetic field within the cavity by applying a low-power (<25 mW) microwave signal at one antenna placed inside. Another antenna measures the response. The resonance frequency is a measure of the permittivity, and thus electron density, of the medium inside.

In this work, however, we frequency modulate the microwave generator with a sawtooth signal at a frequency of 20 Hz. This results in a linear sweep of the microwave frequency over a constant frequency range during each modulation period (50 ms). Concurrently, the cavity response is sampled at 1 kHz, resulting in 50 measurements of the cavity response each modulation period. So, within one modulation period, each cavity response corresponds with a slightly different excitation frequency. If the cavity response values are plotted against this frequency, the characteristic resonance peak (analogous to figure 5 in [37]) is recovered. The peak is fitted for all modulation periods and the position of the maximum is taken as the resonance frequency. Ultimately, this yields the electron density by

$$\bar{n}_e = \frac{8\pi^2 m_e \epsilon_0 f^2}{e^2} \frac{\Delta f}{f_0}, \quad (1)$$

with 50 ms time resolution during one experiment. Symbols have the following meaning: m_e electron mass, e electron charge, ϵ_0 permittivity of free space, f the resonance frequency with plasma, f_0 the resonance frequency without plasma/in vacuum, and $\Delta f \equiv f - f_0$. In previous investigations, multiple experiments were needed to (re-)construct the picture in frequency space. This greatly limited the experiment duration that could be probed, since experiments start to deviate from one another already after several seconds. Using the modulation method, however, one considers a single dusty plasma experiment. Reproducibility is therefore not an issue. The only drawback is a reduced temporal resolution. However, 50 ms proved adequate for the dynamics we were interested in. As a result, we were able to determine the electron density over a period exceeding minutes of one dusty plasma experiment with non-intrusive MCRS.

As mentioned before, the principle of MCRS is that a standing wave of the electromagnetic field is excited inside the cavity. The standing wave will oscillate at the excitation frequency. Together, the nodal pattern and resonance frequency of the standing wave are known as resonant mode. For a simple cavity geometry, the resonance frequencies and mode patterns can be calculated [38]. For an ideal cylindrical cavity, two types of modes can exist: transverse electric (TE) and transverse magnetic (TM). Each resonant mode is symbolized by subscripting the type (TM or TE) by the values of three indices: m , n and p . Figure 2(a) depicts the mode pattern for TM_{030} at a vertical position halfway between the end plates inside a cylindrical cavity (dashed). In this case, the only non-zero component of the electric field is the one in the vertical direction z . Moreover, the mode is height-independent. This allows us to visualize the electric field value by a displacement in the vertical direction. More specifically, the red ring coinciding with the circumference of the cavity has a value of zero, whereas the central yellow-colored peak has a high positive value.

A property of MCRS is that the electron density is weighted by the squared electric field magnitude of the corresponding resonant mode,

Figure 2. (a) Mode pattern (electric field) for TM₀₃₀ shown at one of its extremes. Half a period later, the pattern will be reversed with respect to the vertical direction. (b) Normalized squared electric field (E^2) for TM₀₃₀ as a function of radial position in the cavity. An electron density $\bar{n}_e$ determined using this mode will be weighted by E^2 . For clarity, the corresponding sensitivity to free electrons in the cavity is also indicated in this graph.

$$\bar{n}_e = \frac{\int n_e(\vec{x}) E^2(\vec{x}) d\vec{x}}{\int E^2(\vec{x}) d\vec{x}}, \quad (2)$$

with $n_e(\vec{x})$ the electron density and $E(\vec{x})$ the electric field magnitude of the resonant mode at a point $\vec{x}$ in space, where the integrals are evaluated over the total cavity volume. In short, electrons are probed most where E^2 is maximal. For the mode depicted in figure 2(a), E^2 is plotted as a function of radial position in the cavity in figure 2(b). This mode is mostly sensitive to electrons close to the central axis of the cavity, whereas it will not measure any free electrons present at the circumferential wall (and anywhere else where $E^2 = 0$). It is moderately sensitive to electrons at several positions between the central axis and the circumferential wall as indicated in the figure.

In this work, two resonant modes are employed to provide a better picture of where changes in the electron density occur. To this end, the actual field patterns inside the cavity are determined by locally and systematically disturbing the medium (without plasma) with a small PVC cube (10 mm × 10 mm × 5 mm) suspended by a thin (Ø 0.3 mm) nylon string. For more details the reader is referred to [39], which extensively describes this method.

The suspended dust cloud is visualized using laser light scattering. To that end, a laser beam emitted by a laser diode (532 nm, 20 mW CW) is converted to a (vertical) light plane of about 0.5 mm thickness with the help of two collimating lenses and a cylindrical lens. The light plane enters the vessel through a small port. A plain mirror guides the light plane through a slit in the discharge chamber's bottom lid (see above), after which it reaches the top lid (powered electrode). The intensity of the scattered laser light is a function of several experimental parameters, of which dust particle size, density and refractive index are most important. Even if the refractive index is assumed constant over the growth cycle of the

particles (which is questionable), it is with the present setup impossible to decouple particle size and density from a change in scattering intensity. Ellipsometric techniques could resolve this issue as recently published by Greiner *et al* [40]. It should be noted that scattering intensity varies much more strongly with particle radius (in the Rayleigh regime with r^6) than it does with particle density (linear).

The light coming from the discharge volume (containing both scattering and plasma emission) exits the vessel through one of its main ports. A beam splitter splits this light in two equal parts, with each path being recorded by one CMOS camera. The optical path is approximately 1 m and as such minimizes perspective distortion. One camera employs a dielectric optical filter with a 10 nm FWHM (full width at half maximum) window centered on 759 nm. The transmission of the filter was measured using a halogen lamp and an Ocean Optics HR4000 spectrometer. The plasma emission of a dusty argon–acetylene discharge was obtained using an Ocean Optics HR2000+ spectrometer. Both spectra are shown in figure 3. The filter is roughly 5 times more sensitive to the 763.51 nm transition of Ar I than it is to the ones around 750 nm. Other atomic transitions are negligible. We selected the 763.51 nm line because it is fairly isolated within the spectrum. Analogously, the other camera records light centered on the laser diode wavelength of 532 nm (same FWHM). In this spectral region the plasma itself shows no line radiation and the continuum radiation is negligible compared to the scattered laser light intensity (confirmed with an Ocean Optics HR2000+ spectrometer). After the dielectric filter, the light path is focused on the CMOS chip using a $f = 25$ mm collimating lens.

The 763.51 nm line is a transition towards a metastable state of argon. This means that the photons can be subject to re-absorption, an effect that is enhanced if the metastable density increases. Such an increase in metastable density has actually been observed in dusty plasmas before [41]. The transition at 750.39 nm does not suffer from this problem.

Figure 3. Plasma emission intensity (blue) of a dusty argon–acetylene plasma and the transmission of the optical filter (red). The optical filter is roughly 5 times more sensitive to the 763.51 nm transition than to the ones around 750 nm. Other atomic transitions are negligible.

However, under our experimental conditions, the emission from 763.51 nm does not show significant re-absorption. We verified this by collecting the plasma emission from the center of the discharge using an Ocean Optics HR2000+ spectrometer. The ratio of the intensity from the lines at 763.51 nm and 750.39 nm stays constant during the whole dusty plasma experiment, as shown in figure 4. This means that the line at 763.51 nm behaves similarly as the line at 750.39 nm and it is not significantly influenced by re-absorption.

The cameras are synchronized with each other by triggering them externally using a pulse generator set at 10 Hz. The signal from the pulse generator is also fed to a second channel of the data acquisition system responsible for recording the electron density. This creates a common timebase between all the temporally resolved measurements.

3. Observation

After switching on the plasma, light scattering becomes visible after about 20 s. At that time a small void is already discernible. In the following moments the void expands in a steady fashion and the light scattering of the dust particles becomes more intense. This is designated phase *I*. At some moment in time, the void rapidly expands (phase *II*), while external conditions are not changed. The expansion stops and the void contracts a small amount: the ‘hiccup’ (phase *III*). The void then continues its expansion (phase *IV*). During phase *IV* a scattering signal starts appearing from within the void, indicative of a new batch of dust particles that have formed within the void. At this point the formation of a new void also becomes visible and the situation is identical to the start of phase *I*. The whole process repeats itself until the plasma is switched off. The process is illustrated in figure 5. A video file is supplied as supplementary material (stacks.iop.org/JPhysD/48/035204/mmedia).

Figure 4. Ratio of plasma emission of the transition at 763.51 and at 750.39 nm ($I_{763.51}/I_{750.39}$) for a dusty argon–acetylene experiment (blue). The electron density as measured using MCRS is also shown (red). It is clear that both transitions behave similarly.

The scattering intensity along the vertical dashed line in figure 5 is plotted as a function of time in figure 6. All the phases *I* through *IV* are also clearly visible within this figure. Close examination of the figure reveals that during the phase *III* (the hiccups) the scattering intensity (most visibly within the void region) briefly goes up.

The size of the void (two-dimensional cross-sectional area) was tracked using a Canny edge detection algorithm in Matlab. The result is presented in figure 7(a) overlaid with the weighted electron density as measured with MCRS. The plasma is switched on at 0 s. The algorithm starts to detect a void at about 37 s. At about 231 s the plasma is switched off. For this particular experiment, four cycles of void formation and expansion have occurred and would have continued had the power not been switched off. The four cycles, and within them the expansion hiccups are clearly visible. A zoom of the first cycle around the rapid void expansion and hiccup is shown in figure 7(b). This shows that the void expansion is accompanied by an increase in electron density and the void hiccup by a similar decrease in electron density. The phases as illustrated in figure 5 are also annotated.

A plot of the relative change in emission during the void hiccup is shown in figure 8. Red values indicate a relative increase in emission during the hiccup, whereas blue values signify a relative decrease. The color white signifies no change at all. The dashed curve corresponds to the void size at the onset of the void hiccup. It can be clearly seen that during the hiccup the emission significantly increases predominantly within the void region.

Two other experiments were conducted under the same conditions although measured with different MCRS resonance modes. In the first experiment, mode A in figure 9(a) was used, whereas for the second experiment mode B was used. The resonance mode profiles were obtained as described in the Experimental details section. Figure 9(a) therefore shows the radial distribution of the standing wave pattern for

Figure 5. Illustration of the cyclic behavior of the dust void expansion. The dashed vertical line in panel a is used for figure 6. To guide the eye, the dashed curve in panel c indicates the position of the void edge at the time of panel d.

Figure 6. Scattering intensity of the central region of the discharge (along the vertical dashed line in figure 5). The top of the graph coincides with the upper electrode, the bottom of the graph with the lower electrode. The phases as illustrated in figure 5 are also annotated.

Figure 7. (a) Electron density measured with MCRS during one dusty plasma experiment (left axis) overlaid with the void cross-sectional area (right axis). The plasma is switched on at 0 s and switched off at about 231 s. (b) Rescaled zoom around the first hiccup. The Roman numerals correspond with the phases in figure 5.

two different microwave resonant modes. As described in detail in the Experimental details section and shown in particular in figure 2, a resonant mode present in a resonant cavity

will show a three-dimensional nodal pattern (of the electromagnetic field) specific to the mode in question (TM_{mnp} or TE_{mnp}). In turn, the nodal pattern reflects the sensitivity to free

Figure 8. Relative change in emission of Ar I at predominantly 763.51 nm during the void hiccup. The values are calculated on a per-pixel basis using $(I_2 - I_1)/I_1$, where I_i is the emission intensity at time t_i ($i = 1, 2$) as annotated in figure 7(b). The dashed curve indicates the position of the void boundary at time t_1 . It is seen that during the void hiccup the plasma emission increases substantially and almost exclusively within the void region.

Figure 9. (a) Electric field profile of two resonant modes near 6.2 GHz measured close to the upper electrode and perpendicular to the plane of antennas (along the r -axis in the upper right inset). Analogous to figure 2(b), this figure shows the sensitivity of both modes to free electrons as a function of radial position in the cavity. (b) Electron densities measured with MCRS using both resonance modes. The jumps in electron density during the void hiccup are much less pronounced in the measurement with mode A. Note that these were consecutive measurements so the timing can vary slightly.

electrons (see equation (2)). As a result, the spatial sensitivity to free electrons differs from mode to mode. The (measured) modes A and B presented in figure 9(a) loosely follow each other near the edges of the cavity ($|r| > 4$ cm). However, near the center ($|r| < 4$ cm), mode B exhibits much higher electric field values than mode A. Consequently, compared to mode A, mode B is much more sensitive to free electrons surrounding the central axis of the discharge chamber. Because our plasma chamber is the resonant cavity for our experiments, the resonant modes are excited within the same region as the radio-frequency plasma. The microwaves will, however, not disturb the radio-frequency plasma since the power of the microwaves is much smaller (in excess of a factor 500) than the power

dissipated by the radio-frequency plasma. The resulting electron densities are shown in figure 9(b). With mode A, an electron density is obtained that is fairly flat after the initial period and it does not exhibit the characteristic jumps during the void hiccup. These jumps are present when measuring with mode B, as well as in the measurement presented earlier in figure 7 (which was obtained using a resonance around 7.51 GHz).

4. Model for stages I and II

To get a better understanding of what is happening during the phases leading up to the hiccup, a simple analytical model for

Table 1. Input parameters for calculation of the magnitude of several important forces in our experiment.

Parameter (symbol)	Value
Ion/neutral temperature (T_i, T_n)	300 K
Electron temperature (T_e)	2.5 eV
Ion/neutral (argon) mass (m_i, m_n)	6.64×10^{-26} kg
Thermal conductivity for argon (σ_T)	$0.0177 \text{ W m}^{-1} \text{ K}^{-1}$
Ion, electron density (n_i, n_e)	$5 \times 10^{14} \text{ m}^{-3}$
Neutral gas density (n_n)	$2.41 \times 10^{21} \text{ m}^{-3}$
Neutral gas speed (u_n)	0.15 m s^{-1}
Dust particle mass density (ρ_p)	1.51 g cm^{-3}
Electric field (E)	50 V m^{-1}
Temperature gradient (∇T)	150 K m^{-1}
Ion-neutral collision cross-section (σ_{in})	$8 \times 10^{-19} \text{ m}^2$

the forces acting on a dust particle is solved that regards an isolated dust particle at the void edge in a one-dimensional discharge. At the void edge the most dominant forces are the ion drag force (F_i) and the electric force ($F_E = Q_p E$) resulting from the local (ambipolar) electric field (E) working on the dust particle charge (Q_p). Other forces (gravity, neutral drag, radiation pressure and thermophoresis) are estimated to be at least an order of magnitude smaller than the electric and ion drag force. This was verified using the expressions for the forces given in [42], but are summarized in the appendix A. The input parameters are given in table 1. Dust particle charge was calculated using orbital motion limited theory [43, 44] (neglecting negative ions), where the time to reach charge equilibrium is much smaller (<1 ms for particles bigger than 10 nm in diameter) than characteristic time scales in our experiment (seconds) [45]. An overview of the magnitude of several important forces as function of particle radius is presented in figure 10, from which it indeed follows that under present conditions F_i and F_E are dominant for particles up to several hundred nm.

For particles at a stationary void edge, hereafter referred to as ‘edge particles’, F_i balances F_E . The ion drag force increases roughly quadratically with particle radius, whereas the charge, and therefore electric force, scales linearly ($Q_p = 4\pi \epsilon_0 r_p \Phi_s$, with Φ_s the floating potential at its surface). Growing dust particles experience an increasing ion drag force which pushes the edge particles to a position where, again, $F_E = F_i$. Since the electric field increases towards the sheaths, growing dust particles will be pushed towards the discharge walls and the void will expand.

It is estimated that our dust particles detectably scatter green laser light when they are at least 50 nm in diameter, meaning that they are in the steady growth, or accretion phase (see Introduction). During this phase particles increase in size with an almost constant growth rate of several nm per second, depending on the experimental conditions [17]. To determine the equilibrium position of the edge particle one needs to solve $F_i(x) = F_E(x)$ for the position x . For this, the actual electric field profile of the dusty plasma is needed. In actuality, the plasma potential profile Φ , and therefore the electric field $\mathbf{E} = -\nabla\Phi$, has a rather complex shape. Due to the particle growth, both F_E and F_i will increase. If the rate of increase

of both forces is almost comparable, the equilibrium position will change only slightly. If, however, F_E cannot keep up, as it were, with F_i , the equilibrium position is reached increasingly farther away from the void center. In other words, the void expansion would accelerate. This is what is observed between phase I and phase II.

We consider a representative plasma potential for a comparable low-pressure dusty plasma as presented in figure 7 of [24]. In that publication, the authors developed an extensive self-consistent numerical model for a dusty argon–silane plasma. The potential includes effects from all charged species (electrons, ions and dust particles). This potential varies with the vertical position in the discharge and therefore accurately describes the local potential experienced by a dust particle at a certain vertical position. Using this potential, the one-dimensional electric field has a high gradient close to the void center at $x = 0$ (where however $E = 0$) and decreases to a near-constant value up to the pre-sheath several cm away. Between pre-sheath and electrode it of course increases dramatically (again). In our experiments, the downward void expansion is observed far from the sheath regions. This means we may limit ourselves to positions between void center and pre-sheath. As a representative one-dimensional electric field profile we therefore take an exponential as given in figure 11, where the asymptotic value is taken as 50 V m^{-1} compared to about 200 V m^{-1} in [24], since our RF voltage is lower and our electrode distance is bigger (65 mm). To clarify, this model describes only the electric field up to the pre-sheath. Expressions for F_E and F_i are outlined in the appendix A. The appendix also describes how the equilibrium position of one edge particle is calculated. The equilibrium position is calculated as function of dust particle radius, and therefore also as function of time if linear growth is assumed. For this we take a value of 2 nm s^{-1} , giving a metric particle radius of $r_p(t) = 2 \times 10^{-9}t$, with t in seconds, neglecting the growth phases preceding accretion.

5. Interpretation

The result of the model described in the previous section is given in figure 12. This graph qualitatively corresponds to what is observed during phase I and phase II in figure 7. From this we can conclude that the complex interplay between the ion drag force and the electric force is a likely candidate for what is observed. The slightly different profile can be attributed to our choice of electric field profile, non-linear particle growth or mutual (screened-Coulomb) interaction between the charged dust particles. It is, however, worth noting that despite the relative simplicity of the model and the uncertainty of the input parameters, the model results make sense on a quantitative level as well. In particular, the onset of phase II is calculated to occur for particles about 120–150 nm in diameter, a perfectly reasonable size at this stage of the dust formation process. Transmission electron microscopy of particles collected previously under similar conditions below the center of the discharge had grown to a diameter of about 190 nm after several cycles and were fairly monodisperse, see

Figure 10. Magnitude of five of the most important forces working on a microparticle in a dusty plasma as function of particle radius. See text for conditions.

Figure 11. One-dimensional electric field profile within our discharge used as input for the force balance $F_i(x) = F_E(x)$. A position of 0 mm corresponds with the void center.

figure 13. The position for which the model predicts the fast expansion is around 1 cm, close to what is observed in reality. It takes about 30 s to reach this stage, which is close to what is observed as well. However, going beyond this simple estimate/model is extremely difficult since accurate experimental data (e.g. the potential profile) are lacking. Moreover, a quantitative model would necessarily need to solve the complete system self-consistently, including Boltzmann's equation, Poisson's equation and the dust particle growth and dynamics.

The reason that an increase in electron density is observed during phase II has all to do with a decreasing sink of electrons that an expanding void brings about. Either, the quickly expanding void pushes dust particles at the edge of the chamber out (recall that the walls are made from a metal mesh). During this process the electron density can temporarily go up while the plasma responds to the new situation

of fewer dust particles and therefore lower sink. Or, since the void region itself has a relatively high free-electron density compared to the dusty region [46], a quickly expanding void enlarges the dust-free region and can therefore result in a higher (volume-averaged) free-electron density, as measured using MCRS. In reality, both processes are likely to contribute to the observed increase in weighted electron density.

Phase III is the spontaneous contraction of the expanding dust void, which we refer to as 'hiccup'. We infer the hiccup is caused by coagulation of a new batch of growing dust particles within the expanding void. This reasoning is supported by several observations. During the hiccup we—on the one hand—observe a decrease in electron density, but only when using the microwave resonance mode that is sensitive to electrons in the void region, i.e. close to the cavity's center. On the other hand, an increase in plasma emission intensity (from mostly the 763.51 nm transition) is seen and primarily located within the void region. Also, the laser scattering intensity briefly goes up, indicative of the abrupt size increase of the particles which has a large effect on the scattering intensity (for Rayleigh $\propto r_p^6$). The decrease in electron density is in accord with the general picture that during the (sudden) coagulation phase the electron density goes down due to the (sudden) increase of electron losses on the dust particle surface. The void contracts as a result of a smaller ionization rate causing a decreasing ion density and hence smaller ion drag force.

Unfortunately, MCRS does not yield direct spatial information about the electron density. However, by combining multiple resonant modes and looking at their electric field patterns, it is possible to obtain more information about where changes in the electron density occur. The most striking difference between the electric field patterns of modes A and B in figure 9(a) is that mode A has a node at the center of the discharge chamber, i.e. the mode is insensitive to free electrons in this region. Away from the center, the mode rapidly increases. A (weighted) electron density measured using this

Figure 12. Equilibrium position of one edge particle where the ion drag force balances the electric force as function of particle radius, and when assuming a constant growth rate also as a function of time.

Figure 13. (a) Six transmission electron micrographs (TEM) of the dust particles collected below the center of the cavity on a single TEM grid. The plasma was switched off at the maximum of the third jump in electron density, see figure 7. The scale bar has a length of 50 nm. The flows of argon and acetylene were 16.4 sccm and 3.4 sccm, respectively. The plasma-dissipated power was a bit higher compared to the other measurements in this paper: about 23 W compared to about 19 W. (b) Size distribution of 49 particles. The left group is fitted with a Gaussian, yielding an average size of 185 nm and a standard deviation of about 8 nm.

mode will predominantly be caused by free-electrons in the regions away from the central axis. Mode B loosely follows the profile of mode A, but has a much higher sensitivity near the center. In other words, mode B is sensitive to free electrons near the central axis of the discharge, whereas mode A is not. Consequently, the difference between the magnitude of the jumps in electron density at the void hiccup measured with mode A compared with mode B are most probably caused by free electrons near the central axis of the discharge. Since the void is also positioned around the central axis of the discharge, this is equivalent to saying that the jumps in the electron density are probably due to changes occurring within the void region. This in turn supports the view that it is coagulation within the void region that lies at the heart of

the void expansion hiccup. The increased emission within the void region is due to an increased electron energy that is usually associated with coagulation (see also the Introduction).

The void dynamics could also be influenced by a changing ambipolar electric field during the void's expansion, since inside a void the gas fraction being ionized is higher compared to the dusty part surrounding it. However, this would probably result in a more gradual void response since its expansion is also gradual, at least in the roughly 4 s up to the void hiccup (68–72 s in figure 7(b)). The observed hiccup is, however, rather sudden. This points to a sudden change in forces to be exerted on the particles. A much more likely candidate is therefore coagulation, which is a sudden event as well. Moreover, the hiccup exactly coincides with the slight—and

sudden—drop in electron density in the void (as well as with the increased emission). This also points to a phenomenon that suddenly decreases the electron density in the void, for which (again) coagulation is the logical candidate.

6. Conclusions

We have observed cyclic dust particle formation in a low-pressure argon–acetylene discharge. Within each cycle, a dust-free zone (dust void) develops. Its expansion can be characterized by four phases: a steady expansion, a rapid expansion, a contraction (‘hiccup’), and finally a continued expansion.

A simple analytical model for the two dominant forces (ion drag and electric) qualitatively reproduces the steady and rapid expansion. The complex interplay between these two forces is, therefore, a likely candidate for what is observed. During the void hiccup, the electron density decreases predominantly within the void region, whereas the plasma emission increases there. A small increase in scattering intensity is also observed during the hiccup. Several seconds after the hiccup, light scattering from dust particles within the void becomes visible. These observations point to an abrupt and collective coagulation of a new batch of dust particles growing within the void region at the onset of the hiccup.

Couëdel *et al* [36] also hypothesized that the void instability they observed is due to the formation of a new batch of dust particles in the void. We underscore this reasoning, although the timescales and dynamics of their void instability differs greatly from ours. By employing similar measurement techniques as presented in this Paper, in particular microwave cavity resonance spectroscopy, as well as simple modeling of the void dynamics more light can be shed also on that particular experiment.

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Appendix A. Model details

A.1. Forces

Symbols as defined in table 1.

A.1.1. Gravity.

$$\vec{F}_g = \frac{4}{3}\pi r_p^3 \rho_p \vec{g}. \quad (\text{A.1})$$

A.1.2. Neutral drag force

For the neutral drag force the expression from Epstein (kinetic regime, relative dust particle speed smaller than neutral thermal speed) is used (stationary dust particle),

$$\vec{F}_n = \frac{4}{3}\pi r_p^2 m_n n_n \sqrt{\frac{8k_B T_n}{\pi m_n}} \vec{u}_n. \quad (\text{A.2})$$

A.1.3. Thermophoretic force

For the thermophoretic force the approximation by Talbot is used,

$$\vec{F}_{th} = -\frac{32}{15}r_p^2 \sigma_T \vec{\nabla} T \sqrt{\frac{\pi m_n}{8k_B T_n}}. \quad (\text{A.3})$$

A.1.4. Electric force

From orbital motion limited theory follows a recursive expression for the dust particle floating potential Φ_s ($\Phi_s < 0$)

$$\exp\left(\frac{e\Phi_s}{k_B T_e}\right) = \sqrt{\frac{m_e T_i}{m_i T_e}} \left(1 - \frac{e\Phi_s}{k_B T_i}\right). \quad (\text{A.4})$$

This expression is solved numerically and yields the dust particle charge (in coulomb) by regarding a dust particle as a perfectly spherical capacitor

$$Q_p = 4\pi\epsilon_0 r_p \Phi_s. \quad (\text{A.5})$$

The resulting electric force follows from the charge multiplied by the local electric field,

$$\vec{F}_E = Q_p \vec{E}. \quad (\text{A.6})$$

A.1.5. Ion drag force

The ion drag force contains a so-called ‘collection’ and ‘orbit’ component. The total force is given by

$$\vec{F}_i = \pi n_i m_i \vec{u}_{i,dir} u_i (b_c^2 + 4b_{\pi/2}^2 \Gamma). \quad (\text{A.7})$$

Here,

$$\vec{u}_{i,dir} = \sqrt{\frac{2\vec{E} e \lambda_{mf}}{\pi m_i}} \quad (\text{A.8})$$

is the directed ion velocity for mono-energetic ions as a result of the electric field, with $\lambda_{mf} = 1/(n_n \sigma_{in})$. Furthermore,

$$u_i = \sqrt{u_{i,dir}^2 + \frac{8k_B T_i}{\pi m_i}} \quad (\text{A.9})$$

is the total ion speed and

$$b_c = \sqrt{r_p^2 \left(1 - \frac{2e\Phi_s}{m_i u_i^2}\right)} \quad (\text{A.10})$$

and

$$b_{\pi/2} = r_p \frac{-e\Phi_s}{m_i u_i^2} \quad (\text{A.11})$$

are the impact parameters associated with the collection and orbit component, respectively. Moreover,

$$\Gamma = \ln \left(\frac{\lambda_s + b_{\pi/2}}{r_p + b_{\pi/2}} \right), \quad (\text{A.12})$$

with

$$\lambda_S = \frac{\lambda_{De}\lambda_{Di}}{\sqrt{\lambda_{De}^2 + \lambda_{Di}^2}} \quad (\text{A.13})$$

the linearized Debye length given by, respectively, the electron and ion Debye length

$$\lambda_{De} = \sqrt{\frac{\epsilon_0 k_B T_e}{e^2 n_e}}, \quad (\text{A.14})$$

$$\lambda_{Di} = \sqrt{\frac{\epsilon_0 k_B T_i}{e^2 n_i}}, \quad (\text{A.15})$$

where we have taken $n_i = n_e$ in the void.

A.2. Equilibrium position

The equilibrium position of the particle can be determined by providing a spatial profile of the electric field $E(x)$ making both the electric force and the ion drag force dependent on x . For a given dust particle radius r_p , the equilibrium position is determined by solving $F_i(x) = F_E(x)$ for x . Figure 12 is generated by repeating this for several particle radii.

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