Decoupling of dust cloud and embedding plasma for high electron depletion in nanodusty plasmas

Andreas Petersen (✉ apetersen@physik.uni-kiel.de)

Christian-Albrechts-Universität zu Kiel <https://orcid.org/0000-0002-8620-1027>

Oguz Asnaz

Christian-Albrechts-Universität zu Kiel

Benjamin Tadsen

Christian-Albrechts-Universität zu Kiel <https://orcid.org/0000-0002-2964-8543>

Franco Greiner

Kiel University <https://orcid.org/0000-0002-4828-3770>

Article

Keywords:

Posted Date: January 7th, 2022

DOI: <https://doi.org/10.21203/rs.3.rs-1192899/v1>

License: © ⓘ This work is licensed under a Creative Commons Attribution 4.0 International License.

[Read Full License](#)

Decoupling of dust cloud and embedding plasma for high electron depletion in nanodusty plasmas

Andreas Petersen,^{*} Oguz Han Asnaz, Benjamin Tadsen, and Franko Greiner
Institute of Experimental and Applied Physics, Kiel, Germany

(Dated:)

In recent years nanoparticles (nps) have become key technological products, e.g. as coatings with tunable optical gap in third generation solar cells, as nanocrystals for photonic applications, and as pharmaceutical nanocarriers. In particle sources, that use reactive, nanodusty plasmas, a high dust density changes the properties of the dusty plasma compared to a dust free plasma considerably, as the electron depletion leads to a reduced number of free electrons. This is called the Havnes effect and was central for the understanding of the famous spokes in Saturn's rings. We see here, that it is also important for technological applications. Using self excited dust density waves (DDW) as a diagnostic tool, it is possible for the first time, to completely characterize an argon discharge with embedded amorphous hydrocarbon nps of different size and density. The results show, that electron depletion governs the charge of dust grains, while the size of the particles has only a weak influence. The ion density and electric potential profile are almost independent of both, dust size as well as dust density. This suggests, that the rf generated plasma and the dust cloud coexist and coupling of both is weak.

In the mid eighties of the last century, plasma assisted chip production suffered from drop outs due to generated dust particles. This dilemma was the spark, which established the new research field of (nano)dusty plasmas. While it was first presumed that the particles were sputtered from the source wafers, the surprising result was that nanoparticles (nps) can grow inside of a discharge from molecular precursors [1].

In more recent times, the field has seen a paradigm shift, as plasma created nps have become a valuable product, and research focuses more on the generation processes and properties of the particles themselves.

Under the non-equilibrium conditions of low pressure plasmas [2], with relatively high electron energies (around 50 000 K or 4 eV) and ions as well as neutrals at room temperature, chemical reactions are possible, which can not be observed in classical wet chemistry. Reactive, particle producing plasmas, often noble gas plasmas with an admixture of silane [3], acetylene [4] or methane [5], are central for a wide range of technological applications.

Their use in the technical field ranges from coatings to prime surfaces, for surface protection, for optical purposes [6–8], and even to provide tailored nano dots in solar cell coatings [9, 10]. In the medical field however, nps have become relevant as drug carriers [11, 12]. They are a key feature to stabilize mRNA during application [13]. And in recent developments nps are even considered to deliver drugs into the pulmonary system to treat SARS-CoV2 [14].

Several aspects of the creation of dust particles in a plasma have been thoroughly explored: The plasma chemical reactions [15] which lead to free radicals and their polymerization into clusters [16], the importance of trapped, negatively charged precursors [17] and lastly different phases of the growth mechanism [18, 19].

While there are comprehensive approaches to model interaction of all components of a nanodusty plasma [20, 21], the basic physics of the nanodusty plasmas, which are mandatory to understand the processes in reactive plasmas, is largely unexplored. A detailed knowledge of the charging, an understanding of the confinement of nanoparticles, and changes of the plasma parameters are keys to systematically advance the production processes and to learn about synergistic effects.

Support for the understanding of dusty plasmas at high dust densities comes from a completely different side, from astrophysics. The observation of spokes in the rings of Saturn by the passing Voyager 1 and Voyager 2 satellites in 1979/1980 was one of the trigger events for the new field of physics of dusty plasma. To explain the sporadic appearance and disappearance of the spokes, Goertz and Morfill [22] proposed a model, which is based on meteor impacts on the ring, producing plasma densities high enough to levitate negatively charged, micron sized ice particles. Havnes showed that the created plasmas have high dust density [23]. This results in a depletion of electrons and leads to a reduced dust charge [24].

The key equation to describe the condition of a plasma containing a high dust density is the quasi neutrality equation

$$n_i - (n_e + Zn_d) = 0. \quad (1)$$

In the following, we restrict ourselves to a three component plasma of argon ions with a single positive charge e and density n_i , dust particles with a negative charge of Z elementary charges and density n_d , as well as electrons with density n_e . As seen from Eq. (1), a high density of dust particles leads to a depletion of electrons. The $n_e + Zn_d$ term in Eq. 1 creates the negative charge which has to balance the positive ion charge. This means, that a high negative charge Ze of the dust particles effectively reduces the number of free plasma electrons, leading to an electron depleted plasma.

^{*} apetersen@physik.uni-kiel.de

FIG. 1. Effect of the dust density on the potential Φ of a dusty plasma, when the Havnes parameter P is increased. The plasma potential of the dust free plasma is Φ_p , the floating potential of dust grains Φ_f , and the plasma potential in a dense cloud is Φ_c . Adaption from Ref. [24]

FIG. 2. A 2D image of the scattered light of a laser stripe going from right to left through the center of a discharge chamber. On top and bottom the rf driven electrodes with a diameter of 58 mm are highlighted. Using a short exposure time reveals the existence of self excited dust density waves. The region of the DDW-D in the middle is also marked.

Havnes et al [25] introduced a dimensionless order parameter which describes the depletion of electrons, today called Havnes parameter

$$P = 4\pi\epsilon_0 a \frac{k_B T}{e^2} \frac{n_d}{n_i}, \quad (2)$$

with a the particles radius, T temperature, ϵ_0 vacuum permittivity, and k_B the Boltzmann factor. Fig. 1 gives an intuitive picture of the potential in a plasma when the dust density is increased at constant a and T . For $P < 1$ a "dust in plasma" situation is found. The particles are screened by the surrounding electrons and ions and they charge up to their maximum negative value, leading to deep potential wells. Between these wells the electrical potential Φ recovers to the plasma potential Φ_p of the pristine, dust free plasma. The situation changes when $P \approx 1$, the potential wells of the particles start to overlap and the dusty plasma creates its own "cloud plasma potential" Φ_c which is lower than that of the dust free plasma. For even higher Havnes parameter $P \gg 1$, the potential wells are vanishing. The dust particles are on the same potential, this is the regime of electron depletion.

In the astrophysical situation modeling the electron depleted plasma is based on the assumption, that this plasma is embedded in a dust free plasma which can deliver additional ions needed to account for the dust charging [24]. In Ref. [26] this model was named "Boltzmann distributed ion model" (BIM), as its main assumption is, that the ions inside of the dust cloud are provided by an undisturbed surrounding plasma and are Boltzmann distributed. For a low pressure rf plasma the BIM model can not be used, as there is no surrounding plasma to deliver said ions. Instead the ions are produced by ionizing collisions inside of the dusty plasma. The model suggested by Melzer and Goree [27] takes this internal ion source into account leading to a constant ion density model (CIM) [26].

Already in 1994 Barkan could verify the existence of the Havnes effect in a Q machine plasma [28]. In 2011 Goertz et al. performed a comparative study in a large, thermionic magnetic box plasma and could show that the

Havnes effect is observable and can be described with BIM and CIM in such a plasma [26]. However, both experiments used a setup, where micron sized dust was falling through a plasma, i.e. the experimental situation was not fully stationary.

Until 2015 no experiment existed, that could create and characterize a stationary high density dust cloud in an argon plasma. Two achievements made this possible. The first one was the use of sub-micron particles. For sub-micron particles gravity is only a minor force and the particles can fill the whole plasma volume and form a 3D dust cloud. The second achievement was to use a reactive argon acetylene plasma as the particle source. The plasma chemistry in an argon acetylene plasma provides reactive components as precursors, which polymerize and grow to become spherical and mono disperse particles [3, 4, 29]. To achieve the desired three component plasma consisting only of electrons, argon ions and dust particles, the admixture of acetylene to the feed gas can be stopped. Any hydrocarbon remnants are quickly consumed or flushed from the system.

Fig. 2 shows a dust cloud created with this process. The cylindrical cloud is illuminated by a laser stripe (laser L1 (640.3 nm) in Fig. 8). The scattered laser light creates a 2D image of the cylindrical cloud. At the top and bottom a reflection can be seen in the electrodes. The cloud shows the characteristic dust free region in the center named 'void' and self-excited dust density waves (DDW). The dust density in such a cloud is typically $1 \cdot 10^{13} \text{ m}^{-3}$ at an ion density of $1 \cdot 10^{15} \text{ m}^{-3}$ leading to a Havnes parameter of $P = 10 \dots 50$ [30]. This results in a drastic electron depletion with a dust charge of only $10 \dots 50$ elementary charges. In comparison, a single dust particle of 190 nm radius and $P \ll 1$ would gain up to 1600 elementary charges under assumption of the orbital motion limit (OML) [31]. The determination of the dust size, as well as the dust density and the diagnostic of the dusty plasma is challenging and requires special methods.

To measure the dust density via laser light extinction, the optical properties of the dust particles need to be known. These are determined by means of kinetic Mie scattering [32–34] using the rotating compensator polarimeter shown in Fig. 8. The diagnostic of dust charge,

ion density, and electric field uses an analysis method that is based on the dispersion relation of the DDW [30, 35, 36]. From the wave data recorded by the camera C4, local frequencies and wavenumbers can be calculated, using a Hilbert transform approach. A fitting procedure using the dispersion relation of the DDW gives estimates of dust charge, ion density, and electric field [30]. The modeling takes into account electron depletion and the resulting decrease of particle charge using CIM. Although CIM assumes a constant ion density, the local analysis at each radial position leads to spatially changing ion density, as the wave changes its frequency and wavenumber when it moves through the inhomogeneous plasma (see Fig. 2).

RESULTS

For the experimental campaign presented here, we created a dusty plasma with particle radii of 120 to 160 nm. All discharge parameters are kept constant (base pressure 21 Pa, rf power 8 W). For the five radii the full data of extinction, polarimetry and DDW videos is acquired and analysed by the methods presented in [30, 34, 37].

With the fast camera C4 (see Fig. 8) videos of the dust density wave are taken at 4000 fps containing 1k frames. As an example, Fig. 3(a) shows one snapshot of the video for a particle size of 139 nm. Below a vertical average of the image shows the amplitude of the DDW. The red box marks the region used for Fourier analysis and the vertical dashed line at 30 mm marks the rim of the shielded electrode. Fig. 3(b) shows the instantaneous frequencies and wavenumbers ($\omega(r)$, $k(r)$) determined by Hilbert transform. These ($\omega(r)$, $k(r)$) data sets and the radial dust density profiles are the input data for the so called "Dust density wave diagnostic" (DDW-D) which is described in detail in Ref. [30]. The DDW-D is performed for all particle radii. The dust density waves show a linear, i.e. acoustic dispersion, similar to previous results [30].

In Fig. 4 the radial dust density profiles are presented, which were measured by means of the extinction setup (Laser stripe L1 and cameras C1, C2, C3, see detailed explanation in Methods(ii)). Due to the fact, that the creation of larger particles needs more time and the confinement of the dust inside of the plasma is not perfect (see Fig. 9 of Ref. [38]), the total density of the dust is decreased for large dust particles.

To match our kinetic three particle model of a homogeneous, infinite plasma to a measured data set (ω, k)_{ex}, the models dispersion relation is evaluated for its most unstable mode (ω, k)_m. This mode, exhibiting the highest growth rate, will be observed in the experiment. Evaluation is done separately at each radial position. In Methods(iii) the additional input data needed to do the data analysis is given. Fig. 5 presents the results of the DDW-D for different particle radii. Fig. 5(a) shows the ion density profile. It can be seen, that the dust density

FIG. 3. (a) Image of the wave patterns acquired with Camera C4. The image shows the fluctuations by subtracting the mean intensity over all frames from a single frame. The second part shows the column average in units of cameras least significant bit. The dashed black line denotes the extent of the electrodes shield. (b) Local ω and k of the dust density wave determined by means of Hilbert transform.

FIG. 4. Radial dust density $n_d(r)$ for the different particle radii. The color scheme supports visual comparability, the particle radius is a and the radial position r .

(Fig. 4) and dust size (a -axis in Fig. 4) both change significantly, while the ion density does not change. This is a really surprising finding, as it means, that the ion production rate seems to be unaffected. The plasma production, which mainly bases on the generation of argon ions by means of inelastic collisions of electrons with neutral argon atoms, remains untouched. In addition, an observation of Klindworth [39] could be verified. The radial density profile of a dusty plasma with considerable dust

FIG. 5. Results of the DDW analysis for different particle radii a along the radial direction r : (a) ion density $n_i(r)$, (b) electron density $n_e(r)$, (c) electric potential (for the discussion of a common reference see Methods.(iii)), and (d) dust charge q_d . The color scheme supports visual comparability.

density is convex, in contrast to the density profile in a pristine argon plasma, which is concave.

Fig. 5 (b) shows the severe effect of electron depletion. The electron density lies more than a factor of hundred below the ion density, as the dust particles snatch away nearly all free electrons. As a detail, an increase of the electron density is seen for the largest dust particles (160 nm) at larger radial position, far out of the electrodes. Here, the dust density becomes very small and consequently, the electron density recovers. The plasma potential is one of the plasma parameters, that determines the dust density profile, as it provides the electric forces which lead to the confinement of the nps in the discharge. As the DDW-D determines the radial electric field, the potential $\phi(r) = \int E dr + \phi_0$ can only be determined with respect to a reference point, which we set $\phi_0 = \phi(52 \text{ mm}) = 0 \text{ V}$ for all particle radii.

Similar to the ion density, the plasma potential shows no variation with the change of the dust radius and the dust density, Fig. 5 (c).

FIG. 6. Havnes parameter P deduced from the DDW analysis along the radial direction r for all particle sizes a . The color scheme supports visual comparability.

The dependency of the dust charge on the radial position can be seen in Fig.5(d). The dust charge is extremely small in comparison to a single dust particle in a pristine argon plasma. There are no pronounced radial features for the different particle radii. However, an overall increase of the dust charge is seen for increasing particle size. This increase is correlated with the decrease of the dust density (see Fig. 4), as a higher dust density results in a higher electron depletion at constant ion density.

The effect of the dust, i.e. the effect of its size and its density on the dusty plasma is expressed by the Havnes parameter $P(a, n_d, n_i, T_e)$ (Eq. 2). It is no surprise, that P is radially dependent, see Fig. 6. For all particle sizes $P \gg 1$ is valid, the plasma is in a strongly electron depleted state. Again, only a small region radially outside of the electrode shows a smaller Havnes parameter of $P \approx 4$, as the dust density at the highest dust radius is lower at this distance (lower left corner in Fig. 6).

DISCUSSION

An argon plasma was created at constant rf power of 8 W, containing a nanodust cloud at high dust density (high Havnes parameter). The presented data shows, for the first time, that the plasma's radial density and potential profile remain unchanged when the dust size is varied from 117 nm to 158 nm, while the dust density is changed from $6 \cdot 10^{13} \text{ m}^{-3}$ to $1.8 \cdot 10^{13} \text{ m}^{-3}$. This suggests, that the plasma production by means of ionising collisions of energetic electrons with the argon background, and the dust cloud coexist and are not strongly coupled to each other. The electron depletion determines the specific dust charge at a given dust density. The Havnes parameter provides a direct measure of said depletion. The new findings are supported by a Langmuir probe study of Bilik et al [40], which was done in a reactive silan-argon plasma, containing dust. The Havnes parameter of the data presented in [40] can be determined using Eq. 2 and is around $P = 2$, i.e. P is much smaller than in our experiments. Nevertheless the electron depletion is significant and the results are therefore relevant in comparison to the presented results. Bilik et al. observed that the electron energy probability function (EPPF) changes significantly, when dust is present in the plasma. It changes from a Druyvesteyn function to a Maxwellian, leading to a pronounced increase of the "Maxwell tail" of high energy electrons, responsible for the ionisation. The Maxwell tail doesn't change its shape, when the particles grow over time from 25 nm to 150 nm. This observation perfectly matches our finding, that the background plasma remains unchanged for different dust sizes and dust densities, as it means, that the ion sources inside of the plasma remain unchanged during the growth process. The result of Bilik et al. were underpinned by kinetic modeling of Denysenko et al. [41].

FIG. 7. Side view of the plasma chamber. A generator in series with a match box (MB) and balun drives the electrodes with a rf signal of 13.56 MHz. The balun symmetrizes the signal. Gas is fed in on top and pumped out on the bottom of the chamber. For simplicity only the vertical, expanded laser stripe (laser L1 in Fig.8) is shown. The light scattered from L1 produces an image of a 2D cross section of the cylindrically symmetric dust cloud.

METHODS

In this section the experimental setup, the diagnostics and the data analysis methods are presented.

(i) Experimental setup: The experiments are performed in a parallel plate reactor with cylindrical electrodes of 58 mm diameter and an electrode gap of 30 mm. A side view of the chamber and the rf wiring are shown in Fig. 7. A vertical laser stripe illuminates a 2D cross section of the cylindrically symmetric dust cloud. A camera positioned perpendicular to the laser stripe (out of the paper plane in Fig.7) can be used to monitor the dust cloud. Fig. 2 is an example of a typical dust cloud image made by using a red laser and a fast camera. To start the experiments an argon pressure of 21 Pa is set with a flow controller leading to an argon flow of 8 sccm, the plasma is ignited with a rf power of 8 W and 20 % acetylene is added to the argon flow thereby maintaining a constant total gas flow of 8 sccm.

During the experiments, different diagnostics are used to monitor particle size and density. Fig. 8 shows a top view of said diagnostics. A rotating compensator polarimeter (RCP) monitors the polarisation state of light scattered by the growing dust particles from the red laser beam at 662.6 nm (laser L2 in Fig. 8). A matching band pass filter blocks all other light from reaching the RCP. The vertically expanded red laser L1 (640.3 nm) allows to monitor the dust cloud via camera C3, and is used to perform 1D extinction measurements using the intensity of camera C2 calibrated with the initial intensity measured with camera C1. A horizontal, green Laser beam L3 (532 nm) passes the center of the discharge and videos of

FIG. 8. Top view of the experimental setup including the plasma chamber and all optical diagnostics. Three optical diagnostics are combined. (i) Extinction setup for dust density measurements: Laser L1 (expanded to a vertical stripe) and cameras C1, C2 and C3 (ii) Mie Polarimetry for dust size diagnostic: The scattered light of laser beam L2 is acquired by a rotating compensator polarimeter. (iii) Dust density wave diagnostic: Laser beam L3 and fast camera C4.

the scattered light of laser L3 are taken with the fast camera C4 at appropriate times, to acquire video sequences of the dust density wave. A matching band pass filter in front of camera C4 ensures, that only the scattered light of laser L3 is entering the camera. A single snapshot out of one of these video sequences is shown in Fig. 3(a).

As mentioned, the RCP and camera C3 are equipped with filters which block light with a wavelength of 532 nm.

In the following sections the diagnostics for dust size, dust density and complex plasma parameters are described in detail.

(ii) Diagnostic for dust density and particle size.

The dust particles are created using a reactive argon acetylene plasma. After adding acetylene to the argon plasma, spherical amorphous hydrogenated carbon particles (a:C-H) start to grow from molecular precursors up to sizes of a few hundred nanometers in radius. By switching off the acetylene admixture, the particle growth stops, and after consumption of the remaining precursors a complex plasma, consisting only of electrons, ions and (negatively) charged dust is created. The dust particles are spherically shaped and have a narrow size distribution. The rotating compensator polarimeter (RCP) depicted in Fig. 8 acquires a time series of the polarimetric angles Δ and Ψ as well as the degree of polarisation (DoP). This fully characterizes the polarisation state of the scattered light. From this time series the radius of the dust particles is determined by means of kinetic Mie polarimetry [32, 33] and is described in detail in [34]. Fig-

FIG. 9. Kinetic $\Delta(\Psi)$ -Curve (filled circles). The measured values of Δ and Ψ change over time (see colorbar), as the particles size increases. The black curve is the fit of the Mie model. For larger particles a deviation of the model, due to multiscattering, is expected [34]. The "clouds" of symbols along the curve show the settling of Δ and Ψ values after acetylene admixture was stopped. The inset is the SEM image of a typical a:C:H-particle.

ure 9 shows the measured $\Delta(\Psi)$ -curve for a long growth period, the Mie model curve fitted by the algorithm presented in [34], and Δ and Ψ of the aborted growth processes, that create dusty plasmas with differing particle sizes. The outcome of the fitting procedure are the re-

FIG. 10. Dust density profile for a dust radius of $a = 140$ nm.

fractive index of the particles ($N = 2.05 + i0.07$) as well as the radius. The extinction efficiency Q_{ext} of the particles, which is a function of refractive index and particle size, is needed to determine the absolute dust density, from a combination of a 1D extinction measurement (cameras C1 and C2 in Fig. 8) and a video that is taken under a shallow angle from the dust cloud evolving over time (camera C3 in Fig. 8, a telecentric lens is used). The detailed procedure is presented in [42]. Assuming cylindrical geometry, which is justified by computed tomography (CT) measurements [43], the dust density profile can be reconstructed. As an example, Fig. 10 shows the density profile reconstructed for the particle radius $a = 140$ nm. The radial dust density profiles shown in figure 4 and the dust radius provided here are needed as input for the dust density diagnostic, described in the next section.

(iii) Dust density wave diagnostic. The DDWs are waves in a weakly coupled system, i.e. the dust particles are in a gaseous state where the kinetic dust temperature is high compared to the particle-particle interaction. DDWs are ion-dust streaming instabilities of the Buneman type [44, 45]. In the context of dusty plasmas, they were first described in a theory paper of Rao et al. [35]. In 1997 Kortshagen proposed the DDW as a diagnostic for the size of nanoparticles [46]. He used a three-fluid model for dust, ions and electrons in a homogeneous, infinite plasma, assuming that the phase velocity of the DDW is small compared to the ion and electron thermal velocity. The relatively simple fluid model is applicable to our experimental situation, except that the ions have to be treated kinetically as the ion drift velocity is in the range of a few ion thermal velocities [36].

Using experiment related assumptions and simplifications [30] and performing a normal mode analysis of the momentum transfer equations of the three species for small perturbations around the equilibrium yields the dispersion relation of the fluid model

$$0 = 1 + X_e + X_i + X_d \quad (3)$$

$$0 = 1 + \frac{1}{k^2 \lambda_{De}^2} + \frac{\omega_{pi}^2}{k^2 v_{Ti}^2 + \mathbf{k} \cdot \mathbf{v}_i (-\mathbf{k} \cdot \mathbf{v}_i + i\nu_{in})} + \frac{\omega_{pd}^2}{k^2 v_{Td}^2 - \omega (\omega + i\nu_{dn})} \quad (4)$$

with k, ω the wave vector and angular frequency, λ_{De} the electron Debye length, v_{Ti}, v_{Td} the thermal speed of the ions and the dust, ω_{pi}, ω_{pd} their plasma frequencies, and ν_{in}, ν_{dn} the ion-neutral and dust-neutral collision frequencies. Equation (4) has the analytical solution

$$\omega = -i\frac{\nu_{dn}}{2} + \sqrt{-\frac{\nu_{dn}^2}{4} + k^2 v_{Td}^2 + \frac{\omega_{pd}^2}{1 + \chi_e + \chi_i}}. \quad (5)$$

In this equation the ion term is substituted by a kinetic expression to account for the influence of the wave activity on the thermal motion of the ions [30].

Most of the parameters needed to compute the waves complex frequency ω can be determined from the experiment, yet two parameters remain unknown, the ion density n_i and the ion drift velocity v_{i0} . Both variables are taken to be the free parameters of the model and can be fitted to the (ω, k) pairs measured in the experiment. Through quasi neutrality and drift approximation [47]

$$0 = q_d n_d + e n_i \quad (6)$$

$$v_i = \mu E \quad (7)$$

n_i and v_{i0} are directly related to the electric field E and the dust charge q_d . Because electron depletion is important in this case, the CIM model [27] is applied to take it into account. Fig. 1 shows, that a lack of electrons leads to a reduced floating potential. This floating potential is determined from the "floating condition", i.e. the balance of the ion and the electron charging currents (eq. 8, assuming OML approximation) and the quasi neutrality of the dusty plasma (eq. 9), which are two coupled equations that are solved numerically:

$$0 = \left(\frac{m_i T_e}{m_e T_i} \right)^{-\frac{1}{2}} \left(1 - \frac{e \Phi_f}{k_B T_i} \right) - \frac{n_e}{n_i} \exp \left(\frac{e \Phi_f}{k_B T_e} \right) \quad (8)$$

$$0 = \frac{n_e}{n_i} - 1 - P \frac{e \Phi_f}{k_B T_e} \quad (9)$$

For the one-dimensional data analysis in the radial direction, the radius of the nanoparticles a and their density n_d as well as the local ω and k have to be known from the experiment. The radius of the nps was measured using Mie polarimetry and the dust density by extinction measurements presented in Methods(ii). To determine the local ω and k a video of the wave activity is taken with the high speed (4000 fps) camera C4, see experimental setup (Fig. 8).

In a first step the data - taken along the radial direction, at the marked position in the middle of Fig. 2 - is Hilbert transformed to estimate the local phase information of the wave. The phase information over time is used to obtain radially resolved (ω, k) pairs (compare Fig. 3(b)). For each of these pairs, the most unstable wave of the dispersion relation (Eq. 5) is calculated, by a least square fit that determines a model (ω, k) pair with the maximum growth rate $\text{Im}(\omega)$. The resulting model dispersion of such a pair (ω, k) can be seen in Fig. 11. The fit at each position is completely independent from

FIG. 11. Example of the solution of the dispersion relation Eq. 5 for typical conditions in our experiment. The real part gives the wave frequency and the imaginary part the growth rate of the dust density wave for each specific wavenumber k .

the other positions. The result of the fitting procedure gives the free parameters in the model n_i and v_{i0} as well as the dependent parameters E and q_d and all radially resolved plasma parameters which are found in the underlying model equations. In addition to the ion density n_i and the dust charge q_d these are the electron density n_e and the Havnes parameter P .

As mentioned above, one result of the DDW-D is the ion velocity v_{i0} . But because the plasma potential Φ_p is most interesting considering the confining forces on the nps, we can determine it by integrating the electric field E , and the result is shown in figure 5(c). Yet because the DDW-D cannot provide values for the near dust free region close to the chamber wall, no zero po-

tential is known. Instead, to have a common reference for the different particle sizes, the potential was set zero at $r = 52$ mm.

It should be mentioned, that the ion and electron temperature can not be determined by our method. Due to the high collision rate with the argon background gas it is assumed that the ions have room temperature $T_i = 26$ meV. In accordance with the experimental results of [40] T_e was chosen to be 5 eV, which is 50 % higher than the typical electron temperature of the pristine argon plasma. However, a variation of T_e between 3 eV and 7 eV reveals only a weak dependency of the results on T_e .

DATA AVAILABILITY

The data that support the findings of this study is not publicly available for legal/ethical reasons but available from the corresponding authors on reasonable request.

CODE AVAILABILITY

The Matlab software to analyse the $\Delta(\Psi)$ curves, to determine the dust density from the extinction measurements and video data, and to analysis the dust density waves are in-house developed. The code is not publicly available for legal/ethical reasons but available from the corresponding authors on reasonable request.

REFERENCES

- [1] G. S. Selwyn, J. Singh, and R. S. Bennett. In situ laser diagnostic studies of plasma-generated particulate contamination. *Journal of Vacuum Science & Technology A-Vacuum Surfaces and Films*, 7(4):2758–2765, July 1989. ISSN 0734-2101. doi:10.1116/1.576175.
- [2] A. Piel. *Plasma Physics - An Introduction to Laboratory, Space, and Fusion Plasmas*. Springer, 2nd edition, 2017.
- [3] C. Hollenstein. The physics and chemistry of dusty plasmas. *Plasma Physics and Controlled Fusion*, 42(10):R93, 2000. URL <http://stacks.iop.org/0741-3335/42/i=10/a=201>.
- [4] J. Berndt, E. Kovacevic, I. Stefanovic, O. Stepanovic, S. H. Hong, L. Boufendi, and J. Winter. Some aspects of reactive complex plasmas. *Contrib. Plasma Phys.*, 49(3, Sp. Iss. SI):107–133, 2009. ISSN 0863-1042. doi:10.1002/ctpp.200910016.
- [5] S. H. Hong and J. Winter. Size dependence of optical properties and internal structure of plasma grown carbonaceous nanoparticles studied by in situ rayleigh-mie scattering ellipsometry. *Journal of Applied Physics*, 100(6), September 2006. ISSN 0021-8979. doi:10.1063/1.2338132.
- [6] U.R. Kortshagen. Nonthermal plasma synthesis of nanocrystals: Fundamentals, applications, and future research needs. *Plasma Chemistry and Plasma Processing*, 36(1):73–84, 2016. ISSN 1572-8986. doi:10.1007/s11090-015-9663-4. URL <http://dx.doi.org/10.1007/s11090-015-9663-4>.
- [7] K. Honda and R. Kobayashi. Fabrication of c-rich a-sic semiconductor nanoparticles having variable optical gaps and particle sizes using high-density plasma in localized area. *Electrochemistry*, 88(5):397–406, 2020. ISSN 1344-3542. doi:10.5796/electrochemistry.20-64071.
- [8] K.-Y. Cheng, R. Anthony, U. R. Kortshagen, and R. J. Holmes. High-efficiency silicon nanocrystal light-emitting devices. *Nano Letters*, 11(5):1952–1956, 2011. doi:10.1021/nl2001692. URL <https://doi.org/10.1021/nl2001692>. PMID: 21462935.
- [9] B. Sain and D. Das. Low temperature plasma processing of nc-si/a-sinx:h qd thin films with high carrier mobility and preferred (220) crystal orientation: a promising material for third generation solar cells. *RSC Adv.*,

- 4:36929–36939, 2014. doi:10.1039/C4RA04610B. URL <http://dx.doi.org/10.1039/C4RA04610B>.
- [10] S.-W. Park, J.-S. Jung, K.-S. Kim, K.-H. Kim, and N.-M. Hwang. Effect of bias applied to the substrate on the low temperature growth of silicon epitaxial films during rf-pecvd. *Crystal Growth & Design*, 18(10):5816–5823, OCT 2018. ISSN 1528-7483. doi: 10.1021/acs.cgd.8b00384.
- [11] M. Santos, P. L. Michael, E. C. Filipe, A. H.P. Chan, J. Hung, R. P. Tan, B. S.L. Lee, M. Huynh, C. Hawkins, A. Waterhouse, M. M. M. Bilek, and S. G. Wise. Plasma synthesis of carbon-based nanocarriers for linker-free immobilization of bioactive cargo. *ACS Applied Nano Materials*, 1(2):580–594, 2018. doi:10.1021/acsanm.7b00086. URL <https://doi.org/10.1021/acsanm.7b00086>.
- [12] P. Michael, Y. T. Lam, E. C. Filipe, R. P. Tan, A. H. P. Chan, B. S. L. Lee, N. Feng, J. Hung, T. R. Cox, M. Santos, and S. G. Wise. Plasma polymerized nanoparticles effectively deliver dual sirna and drug therapy in vivo. *Scientific Reports*, 10(1):12836, Jul 2020. ISSN 2045-2322. doi:10.1038/s41598-020-69591-x. URL <https://doi.org/10.1038/s41598-020-69591-x>.
- [13] R. Medhi, P. Srinoi, N. Ngo, H.-V. Tran, and R. T. Lee. Nanoparticle-based strategies to combat covid-19. *ACS Applied Nano Materials*, 3(9):8557–8580, SEP 25 2020. ISSN 2574-0970. doi:10.1021/acsanm.0c01978.
- [14] M. Rabiei, S. Kashanian, S.S. Samavati, H. Derakhshankhah, S. Jamasb, and S.J.P. McInnes. Characteristics of sars-cov2 that may be useful for nanoparticle pulmonary drug delivery. *Journal of Drug Targeting*, pages 1–11, SEP 25 2021. doi: 10.1080/1061186X.2021.1971236. URL <http://dx.doi.org/10.1080/1061186X.2021.1971236>.
- [15] K. De Bleeker, A. Bogaerts, and W. Goedheer. Modelling of nanoparticle coagulation and transport dynamics in dusty silane discharges. *New Journal of Physics*, 8(9):178, 2006. URL <http://stacks.iop.org/1367-2630/8/i=9/a=178>.
- [16] S. L. Girshick and C. P. Chiu. homogeneous nucleation of particles from the vapor-phase in thermal plasma synthesis. *Plasma Chemistry and Plasma Processing*, 9(3):355–369, SEP 1989. ISSN 0272-4324. doi: 10.1007/BF01083672.
- [17] L. Ravi and S. L. Girshick. Coagulation of nanoparticles in a plasma. *Physical Review E*, 79(2), February 2009. ISSN 1539-3755. doi:10.1103/PhysRevE.79.026408.
- [18] A. Bouchoule and L. Boufendi. Particulate formation and dusty plasma behaviour in argon-silane rf discharge. *Plasma Sources Science and Technology*, 2(3): 204, 1993. URL <http://stacks.iop.org/0963-0252/2/i=3/a=011>.
- [19] E. Kovacevic, J. Berndt, T. Strunskus, and L. Boufendi. Size dependent characteristics of plasma synthesized carbonaceous nanoparticles. *Journal of Applied Physics*, 112(1):013303, 2012. doi:10.1063/1.4731751. URL <https://doi.org/10.1063/1.4731751>.
- [20] P. Agarwal and S. L. Girshick. Sectional modeling of nanoparticle size and charge distributions in dusty plasmas. *Plasma Sources Science & Technology*, 21(5), OCT 2012. ISSN 0963-0252. doi:10.1088/0963-0252/21/5/055023.
- [21] P. Agarwal and S. L. Girshick. Numerical modeling of an rf argon-silane plasma with dust particle nucleation and growth. *Plasma Chem. Plasma Process.*, 34: 489, 2014. doi:10.1007/s11090-013-9511-3. URL <http://dx.doi.org/10.1007/s11090-013-9511-3>.
- [22] C. K. Goertz and G. E. Morfill. A model for the formation of spokes in saturn’s ring. *Icarus*, 53(2):219 – 229, 1983. ISSN 0019-1035. doi:https://doi.org/10.1016/0019-1035(83)90143-4. URL <http://www.sciencedirect.com/science/article/pii/0019103583901434>.
- [23] O. Havnes, G. E. Morfill, and C. K. Goertz. Plasma potential and grain charges in a dust cloud embedded in a plasma. *Journal of Geophysical Research: Space Physics*, 89(A12):10999–11003, 1984. doi:10.1029/JA089A12p10999. URL <https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/JA089A12p10999>.
- [24] C. K. Goertz. Dusty plasmas in the solar system. *Reviews of Geophysics*, 27(2):271–292, 1989. doi: https://doi.org/10.1029/RG027i002p00271. URL <https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/RG027i002p00271>.
- [25] O. Havnes, C. K. Goertz, G. E. Morfill, E. Grün, and W. Ip. Dust charges, cloud potential, and instabilities in a dust cloud embedded in a plasma. *Journal of Geophysical Research: Space Physics*, 92(A3):2281–2287, 1987. doi:10.1029/JA092iA03p02281. URL <https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/JA092iA03p02281>.
- [26] I. Goertz, F. Greiner, and A. Piel. Effects of charge depletion in dusty plasmas. *Phys. Plasmas*, 18:013703, 2011.
- [27] A. Melzer and J. Goree. *Low Temperature Plasmas, Fundamentals, Technologies, and Technique*, volume 1, chapter 6, pages 133–134. Wiley-VCH Verlag, Weinheim, 2 edition, 2008.
- [28] A. Barkan, N. D’Angelo, and R. L. Merlino. Charging of dust grains in a plasma. *Phys. Rev. Lett.*, 73:3093–3096, Dec 1994. doi:10.1103/PhysRevLett.73.3093. URL <https://link.aps.org/doi/10.1103/PhysRevLett.73.3093>.
- [29] A. Bouchoule. *Dusty Plasmas: Physics, Chemistry, and Technological Impact in Plasma Processing*. Wiley, 1nd edition, 1999. URL <https://www.wiley.com/en-us/Dusty+Plasmas%3A+Physics%2C+Chemistry%2C+and+Technological+Impact+in+Plasma+Processing-p-9780471973867>.
- [30] B. Tadsen, F. Greiner, S. Groth, and A. Piel. Self-excited dust-acoustic waves in an electron-depleted nanodusty plasma. *Phys. Plasmas*, 22:113701, 2015. doi: 10.1063/1.4934927.
- [31] J. E. Allen. Probe theory - the orbital motion approach. *Physica Scripta*, 45(5):497–503, May 1992. ISSN 0281-1847. doi:10.1088/0031-8949/45/5/013.
- [32] P.S. Hauge and F.H. Dill. A rotating-compensator fourier ellipsometer. *Optics Communications*, 14(4):431 – 437, 1975. ISSN 0030-4018. doi: [http://dx.doi.org/10.1016/0030-4018\(75\)90012-7](http://dx.doi.org/10.1016/0030-4018(75)90012-7). URL <http://www.sciencedirect.com/science/article/pii/0030401875900127>.
- [33] G. Gebauer and J. Winter. In situ nanoparticle diagnostics by multi-wavelength rayleigh-mie scattering ellipsometry. *New Journal of Physics*, 5(1):38, 2003. URL <http://stacks.iop.org/1367-2630/5/i=1/a=338>.
- [34] S. Groth, F. Greiner, B. Tadsen, and A. Piel. Kinetic mie ellipsometry to determine the time-resolved particle growth in nanodusty plasmas. *Journal of Physics D Applied Physics*, 48:465203, November 2015. doi:

- 10.1088/0022-3727/48/46/465203.
- [35] N. N. Rao, P. K. Shukla, and M. Y. Yu. Dust-acoustic waves in dusty plasmas. *Planet. Space Sci.*, 38(4): 543 – 546, 1990. ISSN 0032-0633. doi:10.1016/0032-0633(90)90147-I. URL <http://www.sciencedirect.com/science/article/pii/003206339090147I>.
- [36] M. Rosenberg. Ion-dust streaming instability in processing plasmas. *Journal of Vacuum Science & Technology A*, 14(2):631–633, 1996. doi:10.1116/1.580157. URL <https://doi.org/10.1116/1.580157>.
- [37] F. Greiner, A. Melzer, B. Tadsen, S. Groth, C. Killer, F. Kirchschlager, F. Wieben, I. Pilch, H. Krüger, D. Block, A. Piel, and S. Wolf. Diagnostics and characterization of nanodust and nanodusty plasmas. *The European Physical Journal D*, 72(5):81, 5 2018. ISSN 1434-6079. doi:10.1140/epjd/e2017-80400-7. URL <https://doi.org/10.1140/epjd/e2017-80400-7>.
- [38] B. Tadsen, F. Greiner, and A. Piel. Preparation of magnetized nanodusty plasmas in a radio frequency-driven parallel-plate reactor. *Phys. Plasmas*, 21:103704, 2014. doi:10.1063/1.4897169.
- [39] M. Klindworth, O. Arp, and A. Piel. Langmuir probe system for dusty plasmas under microgravity. *Rev. Sci. Instrum.*, 78(3), March 2007. ISSN 0034-6748. doi:10.1063/1.2714036.
- [40] N. Bilik, R. Anthony, B. A. Merritt, E. S. Aydil, and U. R. Kortshagen. Langmuir probe measurements of electron energy probability functions in dusty plasmas. *Journal of Physics D: Applied Physics*, 48(10):105204, February 2015. doi:10.1088/0022-3727/48/10/105204. URL <https://doi.org/10.1088/0022-3727/48/10/105204>.
- [41] I. Denysenko, K. Ostrikov, M. Y. Yu, and N. A. Azarenkov. Behavior of the electron temperature in nonuniform complex plasmas. *Phys. Rev. E*, 74: 036402, Sep 2006. doi:10.1103/PhysRevE.74.036402. URL <https://link.aps.org/doi/10.1103/PhysRevE.74.036402>.
- [42] S. Groth, F. Greiner, and A. Piel. Spatio-temporally resolved investigations of layered particle growth in a reactive argon-acetylene plasma. *Plasma Sources Science and Technology*, 28(11):115016, November 2019. doi:10.1088/1361-6595/ab5412. URL <https://doi.org/10.1088/1361-6595/ab5412>.
- [43] C. Killer, F. Greiner, S. Groth, B. Tadsen, and A. Melzer. Long-term spatio-temporal evolution of the dust distribution in dusty argon rf plasmas. *Plasma Sources Science and Technology*, 25(5):055004, 2016. doi:10.1088/0963-0252/25/5/055004. URL <http://stacks.iop.org/0963-0252/25/i=5/a=055004>.
- [44] O. Buneman. Instability, turbulence, and conductivity in current-carrying plasma. *Phys. Rev. Lett.*, 1:8–9, 07 1958. doi:10.1103/PhysRevLett.1.8. URL <https://link.aps.org/doi/10.1103/PhysRevLett.1.8>.
- [45] O. Buneman. Instability, turbulence, and conductivity in current-carrying plasma. *Phys. Rev. Lett.*, 1:119, 08 1958. doi:10.1103/PhysRevLett.1.119. URL <https://link.aps.org/doi/10.1103/PhysRevLett.1.119>.
- [46] U. R. Kortshagen. On the use of dust plasma acoustic waves for the diagnostic of nanometer-sized contaminant particles in plasmas. *Applied Physics Letters*, 71(2):208–210, 1997. doi:10.1063/1.119502. URL <https://doi.org/10.1063/1.119502>.
- [47] L. S. Frost. Effect of variable ionic mobility on ambipolar diffusion. *Phys. Rev.*, 105(2):354–356, January 1957. doi:10.1103/PhysRev.105.354.

ACKNOWLEDGEMENTS

We gratefully acknowledge funding of the project GR1608/8-1 “Dusty plasmas with high electron depletion: Investigation of fundamental mechanisms and properties through particle and plasma diagnostic” by Deutsche Forschungsgemeinschaft (DFG).

AUTHOR CONTRIBUTIONS

F.G. supervised and conceived the project and worked out further plans with A.P., O.A. and B.T.. Author A.P. and F.G. carried out the experiments. A.P., O.A. and B.T. developed and enhanced the software of the DDW-analysis. A.P. did the extinction data analysis and prepared the figures. F.G. and A.P. wrote the manuscript. The results and the manuscript were discussed with all authors.

COMPETING INTERESTS

The authors declare no competing interests.

ADDITIONAL INFORMATION

Correspondence and requests for materials should be addressed to A.P. or F.G.