Dynamic light scattering study of phase transitions in three-dimensional complex plasmas

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Abstract. The phase transitions of a three-dimensional complex (dusty) plasma under gravity are studied by means of the dynamic light scattering technique. The thermal energy of the dust component is determined during the phase transition to characterize the phase state. Detailed parametric studies are performed to examine the impact of the main plasma parameters on phase transitions. The results are found to be in good agreement with the predictions of a theoretical description of phase transitions that considers plasma instabilities to be the main heating mechanism. The distribution of energy in the system is strongly anisotropic. In the melted state, the dust particle movement in the vertical direction has much higher thermal energies than in the horizontal plane. The ratio of the two energies is constant and in particular independent of all the plasma parameters studied. Furthermore, the phase transition of the dust component is a gradual process. A transition front moves through the system in the vertical direction until the whole system reaches the new phase state. The velocities of energy fronts for the horizontal and vertical components of particle motion are very different. In particular, the melted state is a strongly inhomogeneous and anisotropic state in terms of energy transport.
1. Introduction

The term complex or dusty plasma denotes a plasma that contains an additional charged component of macroscopic solid dust particles with nano or micron size. There are different possible mechanisms responsible for the charging of the dust grains, but in most laboratory experiments the fluxes of plasma species such as electrons and ions onto the dust particle surface are leading to a negative charge. The dust grains typically acquire of the order of $10^3–10^5$ elementary charges. In contrast to the other plasma components, the dust particles are strongly coupled and reveal phenomena of self-ordering. Such ordered dust particle systems, also called ‘plasma crystals’, were discovered for the first time in 1994 by Chu and Lin [1], Hayashi and Tachibana [2] and Thomas et al [3].

In this work, micron-sized dust particles are confined in the plasma-wall sheath of a low-temperature argon discharge. Two-dimensional crystals consisting of a single layer as well as three-dimensional crystal structures of hundreds of layers can be realized.

Complex plasmas are a remarkable topic for studying the ordering processes and transport mechanisms of a strongly coupled plasma component. In particular, complex plasmas offer fundamental insight into the mechanisms of phase transitions. The dust particle lattice can be melted by the variation of discharge parameters such as discharge power or gas pressure.

The melted or gaseous phase state is characterized by an unexpected high thermal energy of the dust component, that is, about four orders of magnitude higher compared to the ordered state. Debate continues about the origin of these high energies. The heating contributions due to spatial inhomogeneities [4–6], finite charging time [7] or stochastic charge fluctuations [8, 9] have been found to be of only minor importance.
Another approach concerns the anisotropy of the plasma-wall sheath and related plasma instabilities. The openness of the system, i.e. the continuous energy transfer from the plasma environment to the dust component, results in an ion two-stream instability [10–12] and a phonon instability [13, 14], which are triggering the phase transitions and leading to the enormous thermal particle energy.

The study of phase transition phenomena is quite demanding, and so far such studies have been carried out mostly on dust systems with only a few layers [13–16].

The detailed picture of the phase transitions of a dust system with a number of layers (three dimensional) is as yet unknown. As will be shown, the phase transition reveals another phenomenon that is related to the anisotropy of the plasma-wall sheath. The phase transition does not occur at the same time everywhere in the dust system, but a gradual transition is seen, i.e. a transition front moves vertically through the system.

The standard imaging diagnostics in the field of complex plasmas such as the standard Charged Coupled Device (CCD) imaging, stereoscopy or digital holography have limitations in their temporal and spatial resolution [17]. In particular, examination of the highly energetic gaseous phase state is difficult with these methods. To overcome these limitations, the ‘dynamic light scattering’ (DLS) technique is applied in this work.

DLS is well established in different research fields such as biology, chemistry and physics [18–20]. This technique analyzes the Doppler shift of scattered laser light caused by the movement of scatterers to obtain information about their dynamics and physical properties.

Previous studies [21] applied the DLS method on dusty plasmas to obtain diffusion constants and to determine particle sizes of nano-scaled dust particles. Hurd and Ho [22] observed free (ballistic) particle motion for polydisperse dust particles with a diameter below 200 nm with a Maxwell–Boltzmann velocity distribution.

Khodataev et al [23] studied diffusion constants in a thermal complex plasma with polydisperse dust grains with diameters of the order of 1 µm. Here, the dust particle system showed a comparatively strong coupling, related to a liquid phase state.

However, far too little attention has been paid to the experimental study of phase transitions and the related heating mechanisms that lead to the unexpected high thermal energy of the dust particles in the disordered phase state. In this paper, the DLS technique is applied to three-dimensional complex plasmas consisting of micron-sized, monodisperse dust grains in a low-temperature, radio frequency (RF) discharge.

The first section of this paper will introduce briefly the DLS method. Section 2 gives a thorough review on the literature concerning the ordering and phase transitions of plasma crystals. Section 3 describes the experimental realization of the complex plasma experiment and DLS setup. In section 4, detailed studies of phase transitions under the variation of fundamental plasma parameters are presented. The impact of the anisotropic plasma environment in the plasma-wall sheath on the growth and transport of thermal energy in different directions during a phase transition is examined.

2. Principles of dynamic light scattering

DLS, also known as photon correlation spectroscopy, is a method where the Doppler shift of scattered light, caused by the movement of scatterers, is measured. The spectrum of the scattered laser light reveals a Doppler broadened line, and the width and shape of the line contain information about the dynamics of the scatterers.
The slow particle motion leads to a line broadening ranging between 1 and 100 MHz. In most cases, this cannot be resolved by spectroscopic methods using prisms or gratings. A temporal analysis of the scattered light can be used instead. The temporal fluctuation of the scattered intensity is measured by means of fast photo-detectors and analyzed using time-correlation functions.

The time-autocorrelation function, the correlation of the stochastic signal with itself, reveals the characteristic decay time of the fluctuation and the decay form gives information about the kind of particle motion that is responsible for the fluctuations. The time-autocorrelation function of a function $A(q, t)$ is defined as

$$
\langle A(q, 0)A(q, \tau) \rangle := \lim_{T \to \infty} \frac{1}{T} \int A(q, t)A(q, t + \tau)dt,
$$

where $q$ depicts the scattering vector, $t$ is the time, $\tau$ is the time shift or lag time and $T$ is the duration of the measurement.

There are two major approaches in DLS experiments. In the heterodyne method, a portion of unscattered light $E_L$ (a local oscillator) is superimposed on the scattered light, whereas the homodyne method detects only the scattered laser light.

In both cases, a square-law detector such as a photomultiplier is used, so that the detected signal is proportional to the square of the electric field $s(q, t) \propto |E_D(q, t)|^2$. The time-autocorrelation function of the detector signal $s(q, t)$ is related to the detected electric field $E_D(q, t)$ via

$$
\langle s(q, 0)s(q, \tau) \rangle \propto \langle |E_D(q, 0)|^2 |E_D(q, \tau)|^2 \rangle.
$$

This expression is proportional to $F_{\text{hom}}(q, \tau)$ in a homodyne and to $F_{\text{het}}(q, \tau)$ in a heterodyne measurement, which are defined by

$$
F_{\text{het}}(q, \tau) := \langle \Psi^*(q, 0)\Psi(q, \tau) \rangle,
$$

$$
F_{\text{hom}}(q, \tau) := \langle |\Psi^*(q, 0)|^2 |\Psi(q, \tau)|^2 \rangle.
$$

The expression $\Psi(q, t)$ is the sum over all phase factors of the scattered electric field $E_S(q, t)$:

$$
\Psi(q, t) := \sum_{j=0}^{N} \exp(iq \cdot r_j(t)).
$$

Here, the physical properties of the scatterers are assumed to be identical.

Most theoretical models in DLS deal with the heterodyne autocorrelation function $F_{\text{het}}(q, \tau)$. This is connected with the homodyne autocorrelation function via the ‘Siegert relation’ [24]:

$$
F_{\text{hom}}(q, \tau) = |F_{\text{het}}(q, 0)|^2 + |F_{\text{het}}(q, \tau)|^2.
$$

This relation holds only if the scattered field is distributed over the scattering volume according to a Gaussian distribution. This means that the scattering volume can be divided into a large number of subregions and the scattered intensities of these subregions are statistically independent.

The theoretical expression for $F_{\text{het}}(q, \tau)$ can be simplified if the positions of the scattering particles are statistically independent, e.g. neutral molecules, atoms or macroscopic particles in solution or systems that act as a perfect gas.
In this case, the heterodyne scattering function \( F_{\text{het}}(q, \tau) \) can be written as \([18, 19, 25]\)

\[
F_{\text{het}}(q, \tau) = \left\langle \sum_{i=1}^{N} \exp(iq \cdot (r_i(\tau) - r_i(0))) \right\rangle = \langle N \rangle F_S(q, \tau),
\]

(7)

where \( r_i \) are the positions of the scatterers, \( \langle N \rangle \) is the average particle number in the scattering volume and \( \langle \cdot \rangle \) denotes the average. \( F_S(q, \tau) \) is called the self-intermediate scattering function.

If the velocity distribution of the particles follows a Maxwell–Boltzmann distribution, the self-intermediate scattering function becomes \([18]\)

\[
F_S(q, \tau) = \exp \left( -\frac{1}{2} q^2 \langle v_x^2 \rangle \tau^2 \right).
\]

(8)

The quantity determined in a homodyne DLS experiment is the normalized autocorrelation function \( g_{\text{hom}}(q, \tau) \) that is proportional to \( F_S^2(q, \tau) \):

\[
g_{\text{hom}}(q, \tau) := \frac{\langle s(q, 0) s(q, \tau) \rangle}{\langle s(q) \rangle^2} - 1 = \beta_{\text{coh}}^2 F_S^2(q, \tau),
\]

(9)

where \( s(q, t) \) is the signal of the photo-detector and \( s(q) \) is its temporal average. The coherence factor \( \beta_{\text{coh}} \) is smaller than 1 and describes the finite degree of coherence at the detector surface.

With (8) the normalized autocorrelation function of a homodyne experiment becomes

\[
g_{\text{hom}}(q, \tau) = \beta_{\text{coh}}^2 \exp \left( -\frac{1}{2} q^2 \langle v_x^2 \rangle \tau^2 \right)^2 \equiv \beta_{\text{coh}}^2 \exp \left( -\frac{2}{\omega_q} \tau^2 \right),
\]

(10)

where \( \omega_q \) is the full-width of the Gaussian function.

Thus, the one-dimensional average velocity squared can be obtained from the width of the Gaussian \( \omega_q \) via

\[
\langle v_x^2 \rangle = \frac{2}{\omega_q^2} = \frac{1}{2\sigma^2 q^2},
\]

(11)

where \( 2\sigma = \omega_q \) is used in the expression on the left-hand side.

The assumption of a Maxwell–Boltzmann distribution motivates the introduction of the dust particle temperature \( T_d \). If the particle size and mass density are known, as is the case for the experiments presented here, then the thermal energy can be determined. The thermal energy related to the dust movement in one dimension is defined as \( T_{\text{eV}} := 0.5k_B T_d = 0.5m_d \langle v_x^2 \rangle \), where \( k_B \) is the Boltzmann constant and \( m_D \) is the mass of a dust particle.

3. Dust structures and phase transitions in complex plasmas

3.1. Structural ordering of dust particles

The electrostatic interaction between dust particles can be described by the Debye–Hückel potential, although no physical justification for this approximation has been given until now \([26]\):

\[
\phi(r) = \frac{e Z_d}{4\pi \varepsilon_0 r} \exp \left( -\frac{r}{\lambda_D} \right),
\]

(12)

where \( Z_d \) denotes the number of charges on the dust grain and \( \lambda_D \) is the Debye length.
Figure 1. Phase diagram of systems with the Debye–Hückel interaction. The body centered cubic (bcc) and face centered cubic (fcc) crystal structure is observed for strong coupling. Below the melting line (solid line), the system is in a fluid state. The dashed line separates the bcc and fcc structure. This sketch of the phase diagram is based on the observations made by Morfill and Ivlev [26].

The phase state of a system of particles interacting via the Debye–Hückel potential is characterized by two parameters. Firstly, the coupling parameter, that is, the ratio of the potential energy between two particles and the average thermal energy,

$$\Gamma = \frac{Z^2 e^2}{4\pi \epsilon_0 \Delta k_B T_d},$$  \(13\)

where \(\Delta\) denotes the average inter-particle distance. Secondly, the screening parameter, which is defined as the ratio of the inter-particle distance \(\Delta\) and the screening length \(\lambda_D\),

$$\kappa = \frac{\Delta}{\lambda_D}. \quad (14)$$

The phase state, depending on \(\Gamma\) and \(\kappa\), is illustrated in figure 1. Three different phases can be distinguished here. The solid phase at strong coupling divides into bcc structures and fcc structures and for weak coupling the system is in a fluid state.

The boundary between bcc and fcc was determined by Hamaguchi et al [27] and the triple point is at \(\Gamma = 3.47 \times 10^3\) and \(\kappa = 6.9\). The melting line \(\Gamma^*(\kappa)\) is approximated by Vaulina et al [28]:

$$\Gamma^*(\kappa) = 106 \times \frac{\exp(\kappa)}{1 + \kappa + \frac{1}{2}\kappa^2}. \quad (15)$$

The pure Debye–Hückel potential is not a valid approach in the plasma-wall sheath, because the ion flow deforms the potential and a wake field develops. The ions are focused behind the grain and attract dust grains located downstream.

As illustrated in figure 2, the situation can be described by a simplified picture. The ion focus is replaced by a fictitious particle with positive charge under the restriction of non-reciprocal interaction. This means that the lower dust grain is attracted by the fictitious particle,
Figure 2. Three particles with their wake fields (simplified picture) in an ion flux are shown. Other particles downstream are attracted (dark solid arrows) to the ion focus represented by a fictitious positive charge. The particles tend to align into chains. The horizontal component (perpendicular to the ion flux) of the ion drag force (dashed, single-ended arrows) stabilizes the particle chains further.

but there is no force in the opposite direction. The dust grains are aligned into chains due to the attractive force of the fictitious particles. The chains are further stabilized by the horizontal component of the ion drag force.

A more realistic description of the dust–dust interaction requires the determination of the anisotropic dust grain potential. The grain potential can be calculated by applying the linear dielectric response formalism. The electrostatic potential associated with a point-like charge of $e Z_d$ is [26, 29]

$$\phi(r) = \frac{e Z_d}{2\pi^2} \int \frac{d^3k}{k^2} \frac{e^{ikr}}{\epsilon(\omega, k)}, \quad (16)$$

where $\epsilon(\omega, k)$ is the plasma dielectric function.

The wake structure of $\phi(r)$ and its dependence on plasma parameters were analyzed by analytical approximations of $\phi(r)$ in [30–32] and by numerical models in [33–35]. Some of these works neglect Landau and collisional damping of the wake field by choosing a simple form of the plasma dielectric function $\epsilon(\omega, k)$. This gives only a rough picture of the wake formation, because the damping mechanisms are essential for the amplitude and range of the oscillatory wake field.

Lampe [29] used a more sophisticated approach for $\epsilon(\omega, k)$ and found that the potential depends on three parameters: the Mach number $M = u_i/c_s$, where $c_s$ is the ion sound speed, the ratio $T_e/T_i$, which controls Landau damping, and the ratio of the ion mean free path and the screening length $l_i/\lambda_D$, which controls collisional damping.

Figure 3 illustrates the typical form of the potential $\phi(r, z)$ in the presence of streaming ions. The grain position is at $z = 0$ and the ions are streaming from the left to the right. The focusing of the ion flow causes a region with positive potential downstream of the grain. This plot refers to the results of the simulations by Lampe [29] in which the exact data and a more detailed treatment of the anisotropic grain potential can be found.

Upstream of the grain and to the side, $\phi(r)$ is close to the Debye–Hückel form (12) with an effective Debye length $\lambda_{eff}$ that depends on the Mach number and the direction. For subsonic
ion flows, the ions as well as the electrons contribute to the shielding. In the case of supersonic ions, only electrons are responsible for shielding and the screening length is given by $\lambda_{De}$.

Landau damping and collisions cause a rapid decay of the wake field amplitude. Under typical conditions of an RF discharge, the wake field is truncated after the first node (the first positive potential peak).

### 3.2. Melting and condensation transitions of the dust system

Figure 4 illustrates an image sequence of the melting transition in the dust system. A vertical plane was illuminated by a laser sheet and a CCD camera was used to obtain the images. A stable crystal is observed at a sufficiently large discharge pressure as shown in figure 4(a). This image also depicts the arrangement in vertical chains due to the ion focusing. The structure is melted by reducing the discharge pressure. The melting starts at the bottom of the dust system, closer to the lower electrode as shown in figure 4(b). At this pressure, the system reveals both phase states simultaneously. The melted part shows self-excited oscillations in the vertical direction with large amplitude. When the pressure is decreased further, the melting front moves upward. At low pressures, the whole system is in a disordered state as depicted in figure 4(c). The CCD camera system is not capable of following the fast particle motion anymore, and the particle positions cannot be resolved.

A remarkable property of the disordered state is the unexpected high thermal energy of the dust grains. At very low pressures, the dust grain can reach energies of the order of 1000 eV, which corresponds to a thermal velocity of about 11 cm s$^{-1}$ for particles with $3\,\mu$m diameter and a mass density of $\rho_{MF} = 1.51$ g cm$^{-3}$. Several mechanisms are suggested to explain this phenomenon that is denoted as ‘anomalous heating’ in the literature.

The increase of the thermal energy of the dust component by stochastic charge fluctuations, caused by the discreteness of the charging process, can increase the thermal energy of the dust
Figure 4. A sequence of images demonstrates the melting transition in a complex plasma. The standard CCD technique was used to get single shots of a vertical plane in the dust crystal. In (a), the system is in a well-ordered state revealing arrangement in vertical chains due to the ion focusing. In (b), the discharge pressure is reduced and the melting starts at the bottom of the system. In the melted state, the dust grains show self-excited oscillations in the vertical direction. In (c), the whole system is in the disordered state at low pressure.

component of the order of a few eV. Thus, the stochastically fluctuating charges are a minor contribution to the anomalous heating [36].

If the charge depends on the coordinate of the grain, e.g. in inhomogeneous plasma environments, another contribution to anomalous heating becomes important. Because of the spatial variation of the grain charge, the total energy is not conserved in dust–dust collisions and the mean kinetic energy increases with time [4–6]. Another facet of this heating mechanism is given by the finite charging time. The self-excited vertical oscillations (figures 4(b) and (c)) can be enhanced by the delayed charging of the dust grains, i.e. a grain moves faster than the time required to achieve the equilibrium charge determined by the local plasma conditions. The particle gains energy in the electrostatic potential of the plasma-wall sheath, because the charge on the way down is more negative than on the way up [7].

The non-reciprocal interaction between two dust particles via the wake field is another source of anomalous heating. The dynamics of the particles system is studied using a one-dimensional approach by Melzer et al [13]. In this work, the simplified picture assuming fictitious positive charges as described in figure 2 is used. A system of two dust layers is examined, where only the horizontal movement is considered. The coupling between the dust grains is assumed to be linear. The non-reciprocal attractive force is included by a lower coupling constant within the lower crystal layer.

A similar approach is followed by Schweigert et al [14], but with nonlinear interaction using the Debye–Hückel potential. In the studies of Melandso [37], an infinite dust cloud in three dimensions is analyzed.

All these works show that the non-reciprocal particle interaction is not only responsible for the anisotropy of the lattice structure, but also makes the crystal prone to lattice instabilities (phonons). By the reduction of the neutral gas friction, the lattice instabilities grow to

New Journal of Physics 14 (2012) 093036 (http://www.njp.org/)
large amplitudes and induce the melting transition and lead to the strong increase of the thermal energy.

A more comprehensive approach to explaining the anomalous heating caused by the wake field is described by Joyce et al. [10], Ganguli et al. [11] and Lampe et al. [12]. They consider the dynamically shielded potential (16), which is obtained by applying linear response theory.

They show analytically that an ion-grain two-stream instability is present at low pressures and that it is responsible for the anomalous heating of the dust particles. The growth of the instability can be damped by the combined effects of ion–neutral and dust–neutral collisions. An increase of the discharge pressure above a critical value $p_{\text{cond}}$ leads to stabilization of the instability. Thus, the ion two-stream instability not only explains the anomalous heating at low pressures, but also gives the mechanism that triggers the condensation transition to a strongly coupled state. To understand the condensation transition, a closer look at the theory and predictions of ion two-stream instabilities is advisable.

The theoretical picture described in [10–12] only treats the dust grains as particles, whereas the electrons and ions only appear as a dielectric medium. By doing so, the theory and related models simplify, because the very fast time scales of electrons and ions are eliminated. This assumption is based on the weak coupling between electrons or ions and dust grains.

In the melted state, where the dust is only weakly coupled, the dispersion relation can be written as

$$0 = 1 + \frac{1}{k^2 \lambda_{De}^2} - \frac{\omega_d^2}{\omega (\omega + i \nu_d)} - \frac{\omega_i^2}{(\omega - k \cdot u_i) (\omega + k \cdot u_i + i \nu_i)},$$  \hspace{1cm} (17)

where $\lambda_{De}$ is the Debye length, $\omega_d$ and $\omega_i$ denote the dust and ion plasma frequencies, $u_i$ is the ion stream velocity toward the electrode, and $\nu_d$ and $\nu_i$ are the dust–neutral and ion–neutral collision frequencies, respectively.

Numerical solutions of (17) show that all instability modes are stabilized if the ratios $\nu_d/\omega_d$ and $\nu_i/\omega_i$ are large enough. For the specific case of $\nu_d/\omega_d = \nu_i/\omega_i$, a stability condition can be obtained analytically

$$\frac{\nu_d}{\omega_d} = \frac{\nu_i}{\omega_i} > \sqrt{2} - \frac{M \cos \theta}{1 + \omega_d/\omega_i},$$  \hspace{1cm} (18)

where $M \equiv u/c_s$ is the Mach number with $c_s \equiv \sqrt{k_B T_e/m_i}$ is the ion sound speed, $u$ is the ion speed and $\theta$ is the angle between the wave vector $k$ and the ion streaming speed $u_i$.

The collision frequency for dust–neutral collisions $\nu_d$ is given by the Epstein formula [38]

$$\nu_d = \frac{8 \sqrt{2\pi}}{3} \frac{m_n}{m_d} a^2 n_n v_{T_n},$$  \hspace{1cm} (19)

where $m_n$ and $n_n$ are the mass and density of the neutrals, respectively.

The numerical factor $\delta$ depends on how the molecules or atoms are reflected from the surface of the particle. A value of $\delta = 1 + \pi/8$ is recommended by experiments on complex plasmas [39].

Equation (19) shows that the dust–neutral collision frequency is proportional to the discharge pressure $p$. This is also true for the ion–neutral collision frequency $\nu_i$ that can be written as

$$\nu_i = C_i u_i p,$$  \hspace{1cm} (20)
Figure 5. Stability boundary in the $\nu_d/\omega_d$–$\nu_i/\omega_i$ plane. For a given stability boundary, $\nu_d/\omega_d$ and $\nu_i/\omega_i$ have to be large enough to enter the stable region. This can be achieved for example by increasing the discharge pressure. This sketch is based on the results obtained by Joyce et al [10].

where $C_i$ is the ion collision coefficient depending on the ion species. The dust plasma frequency is given by

$$\omega_d = \sqrt{\frac{Z^2_d e^2 n_d}{\epsilon_0 m_d}}, \quad (21)$$

where $Z$, $m_d$ and $n_d$ are the charge number, mass and number density of the dust. The particle mass $m_d$ is known for the dust particles used in this work. The dust charge is proportional to the electron temperature and the particle radius

$$Z \propto T_e a.$$

The ion plasma frequency is given by

$$\omega_i = \sqrt{\frac{e^2 n_i}{\epsilon_0 m_i}}, \quad (22)$$

where $m_i$ is the mass of the ions and $n_i$ is the ion density.

The stabilization of the ion two-stream instability is evaluated by a stability condition such as (18). Figure 5 shows a sketch of the stability condition in the $\nu_d/\omega_d$–$\nu_i/\omega_i$ plane. To enter the stable region the ratios $\nu_i/\omega_i$ and $\nu_d/\omega_d$ have to be sufficiently large, e.g. by choosing a sufficiently high gas pressure. A more detailed discussion of the stability condition and simulations that show stable and unstable states can be found in the work of Joyce et al [10].

Even though the theory strictly applies only to an infinite dust cloud, good qualitative agreement with the theoretical predictions was found in simulations of only six horizontal layers of dust particles [10].

The simulations show that a finite dust cloud condensates at a different critical pressure $p_{\text{cond}}$ because of boundary effects. An interesting aspect is the convective nature of the ion two-stream instability. The extent of dust heating rises with thickness of the dust cloud. This is a significant result regarding real experiments, inasmuch as the thickness of the dust cloud is limited by the sheath width and is therefore affected by external discharge parameters.
Figure 6. Hysteresis loop in the phase transition of a complex plasma. This graph qualitatively reproduces the hysteresis loop observed in simulations by Lampe et al., but with pressure in units of pascal. The average dust particle energy was obtained during a variation of the discharge pressure. The pressure was first increased and a sudden drop of the average energy indicates condensation at a critical pressure $p_{\text{cond}}$. A subsequent decrease of the pressure leads to melting at a critical pressure $p_{\text{melt}}$. The critical pressure for melting $p_{\text{melt}}$ is substantially less than the critical pressure for condensation $p_{\text{cond}}$.

The stabilization of the instability and therefore the condensation of the dust cloud caused by a change of experimental parameters can be now interpreted in terms of the plasma frequencies $\omega_d$ and $\omega_i$, the collision frequencies $v_i$ and $v_d$, the Mach number and the angle $\theta$ between $k$ and $u_i$.

The melting transition has also been analyzed by Joyce et al. [10], Ganguli et al. [11] and Lampe et al. [12]. In contrast to the simplified heuristic models of Melzer and Schweigert as discussed above, they used the correct representation of the wake field potential in the ion flow (16).

Their simulations show that longitudinal, shear and mixed modes can be driven unstable during the melting transition. The mode that initiates the melting transition is believed to be a single shear mode with wave vector $k$ parallel to the ion flow. This means that the crystal layers oscillate in the horizontal plane relative to each other. This phonon instability can be stabilized at high pressures by the dust–neutral collisions ($v_d/\omega_d$) or the ion–neutral collisions ($v_i/\omega_i$) [11].

The phonon instability that grows in the crystal phase has to be distinguished from the ion two-stream instability in the gaseous phase state. There is no reason why the critical pressures where the instabilities are stabilized are the same for both the mechanisms. Indeed, the simulations described in [10–12] show a hysteresis loop, i.e. the critical pressure for melting $p_{\text{melt}}$ is lower than the critical pressure for condensation $p_{\text{cond}}$.

This is illustrated in figure 6. First, the discharge pressure is slowly increased and a condensation transition occurs (right curve), where the average dust energy drops from $10^4$ to 2 eV. This is followed by a decrease of discharge pressure, which causes a melting transition (left curve) ending in $10^4$ eV again. As can be seen, the critical pressures $p_{\text{melt}}$ and $p_{\text{cond}}$ are separated by about 17 Pa. This model was waiting for experimental proof for several years.
Figure 7. A sketch of the argon discharge and scattering geometry. The dust particles are immersed by a vibrating dispenser (9) through a hole in the upper electrode. The dust cloud (6) is confined in the plasma-wall sheath. The horizontal confinement is realized by a potential applied to an inner segment (7) in the lower electrode. The components of the scattering arrangement are: (1) helium–neon laser, (2) optical attenuator (0.5–20% transmission), (3) Glan–Thompson polarizer, (4) quarter-wave plate, (5) laser mirror system, (10) beam dump, (11) collection system (see the text for details), (12) lower photomultiplier (horizontal particle movement), (13) upper photomultiplier (vertical particle movement) and (14) fast data acquisition PCI Express card and personal computer.

4. The experimental setup

Figure 7 gives a sketch of the plasma discharge and the scattering arrangement. A capacitively coupled argon discharge is driven between two plane parallel aluminum plates. To ignite and sustain the discharge, an RF generator with a working frequency of 13.56 MHz is connected to the upper electrode. The discharge power is between 1 and 20 W and the discharge pressure is adjusted between 0.5 and 200 Pa.

The dust particles are immersed in the plasma by a dust dispenser, a small stainless steel cylinder with a metal mesh at the bottom. The cylinder contains dust powder and is placed above a hole in the upper electrode. If the cylinder is shaken by a vibrating motor, the dust falls into the plasma.
The dust particles are spherical Melamine–Formaldehyde (MF) particles with diameters of 3.24 or 7.23 µm. They have a monodisperse size distribution with a statistical deviation of only ±0.09 µm.

The dust particles are confined in the plasma-wall sheath near the lower electrode. The vertical confinement is realized by the electric field in the sheath. For the horizontal confinement, additional electric fields are necessary. This is realized by an inner segment in the lower electrode that is on a higher potential.

The confined dust systems have a horizontal extension of about 10 cm and a height of roughly 2 cm. The inter-particle separation is typically about 400 µm.

The DLS setup is designed to examine the dust particle motion in the horizontal and vertical directions at the same time. In common DLS experiments, this is realized by two independent laser beams.

In this work, a DLS setup with a single incident laser beam is proposed to measure both components. The incident laser beam has circular polarization. The radiation with polarization perpendicular to the electrode is selected to study the particle movement in the horizontal plane and the parallel polarization component is used to analyze the vertical particle movement.

The helium–neon laser used in this work has a wavelength of 632.8 nm and an output power of 21 mW. The laser beam is attenuated down to a few mW, and the linear polarization of the laser beam is changed to circular polarization with the aid of a quarter-wave plate.

Only the particle movement parallel to the scattering vector is detected in a DLS experiment. The lower photomultiplier detects the dust movement in the horizontal plane, and the upper photomultiplier examines the vertical particle motion. The scattering angles θ₁ and θ₂ are of the order of 10° in the experiments.

The scattered light is detected by collection systems that consist of an entrance aperture a₁, an interference filter to block the plasma emission, a Glan–Thompson polarizing prism and an exit aperture a₂. The design of the collection system is essential for maximizing the coherence factor βcoh on the one hand, and to obtain a sufficiently large scattering volume on the other hand. The coherence factor βcoh in these experiments was of the order of 0.5.

The scattered light is detected by two highly sensitive photomultipliers with a quantum efficiency of about 60% at 632.8 nm. The signal is finally acquired by a fast two-channel PCI Express transient recorder and saved with up to 50 MHz with the data streaming directly on a personal computer.

5. Results

In the following subsections, the DLS method is applied to study the phase transitions in three-dimensional complex plasmas. The phase transitions are induced by a change in the discharge pressure. The autocorrelation functions are measured at different pressures, and a fit to a Gaussian function reveals the thermal energy of the dust component $T_{eV}$. Different experimental parameters are varied to examine their impact on the phase transition.

5.1. The transition curve and the modified Boltzmann fit

The curve of the thermal energy of the dust particles during a phase transition shows a characteristic form. A typical measurement of this transition curve $\tilde{T} = \ln(T_{eV})$ is displayed in figure 8 on a logarithmic scale.
A measurement of the thermal energy during a phase transition plotted on a logarithmic scale. The logarithmic data can be fitted by a modified Boltzmann function (23). The parameters for describing the phase transition are the critical pressure $p_c$, the corresponding logarithmic energy $\tilde{T}_c$, the slope at the critical pressure $\tilde{T}'$, the decay constant at low pressures $c$ and the maximum and minimum logarithmic energies $\tilde{T}_{\text{max}}$ and $\tilde{T}_{\text{min}}$.

The main characteristic features of the curve are a linear decrease of the thermal energy at low pressures and a sudden decay at the critical pressure down to energies that correspond to room temperature.

In order to compare a large number of transition curves quantitatively and thus to render a systematical study of phase transitions feasible, the curve has to be parameterized by an appropriate fit function.

The ‘modified Boltzmann’ function is introduced here as a valid function to fit the transition curves:

$$\tilde{T}(p) = \tilde{T}_{\text{min}} + \frac{\tilde{T}_{\text{max}} - \tilde{T}_{\text{min}} + cp}{1 + \exp \left( \frac{p - p_c}{dp} \right)}.$$  \hspace{1cm} (23)

Note that logarithmic temperatures are used here: $\tilde{T} = \ln(T_{eV})$. The difference compared to an ordinary Boltzmann function is the linear decrease of $\tilde{T}$ at low pressures. In this part, the pure Boltzmann function stays at a constant level. The linear part is considered by the additional expression $cp$ in (23), where $c$ describes the decay rate of $\tilde{T}$. $\tilde{T}_{\text{min}}$ and $\tilde{T}_{\text{max}}$ represent the minimum and maximum logarithmic energies, $p_c$ defines the critical pressure of the transition and $dp$ gives the decay constant at the critical pressure.

The logarithm of the thermal energy at the critical pressure $\tilde{T}_c$ is given by

$$\tilde{T}_c = \tilde{T} |_{p=p_c} = \frac{\tilde{T}_{\text{max}} + \tilde{T}_{\text{min}} + cp_c}{2}.$$  \hspace{1cm} (24)

The slope at this point is expressed by

$$\tilde{T}' |_{p=p_c} = \frac{c}{2} \frac{\tilde{T}_{\text{max}} - \tilde{T}_{\text{min}} + cp_c}{4 dp}.$$  \hspace{1cm} (25)

Note that there is no underlying physical meaning in the choice of this fit function; the analytical form was chosen for convenience.
Figure 9. Cross section of the upper and the lower electrode. The inner segment of the upper electrode is powered with $U_{RF}$. The external bias voltage $U_{dc}$ is applied by a decoupling circuit (dashed box) on the powered segment. The lower electrode has an inner segment to maintain dust confinement. A voltage $U_{C}$ of the order of 20 V is applied to this segment by a multichannel, decoupled dc voltage supply. For the purposes of simplified illustration, the electrodes are drawn closer to each other as they are in the experiment.

A single-step transition like in figure 8 is observed for the melting as well as for the condensing transition. The transitions of the horizontal and vertical components show both the same features, but on different energy scales. The vertical component reaches about one order of magnitude higher energies.

Simulations on a two-layer system show comparable thermal energies during the melting transition for the horizontal particle motion. But, in contradiction to the measurements in this work, a two-step melting transition in the thermal energy is predicted in [14]. In the first step the energy grows but the structural order is still high. In the second step, the spatial order gets lost and the gaseous state is reached.

5.2. The external bias voltage and confinement voltage

Two of the most important experimental parameters for manipulating the dust cloud are the confinement voltage and an external bias voltage.

As shown in figure 9, the external bias voltage is directly applied to the driven electrode by a decoupling circuit. Hence, no self-bias potential can evolve freely, but the electrode potential is fixed. An external dc voltage up to ±50 V can be applied for the common plasma parameters in this experiment. The plasma cannot be operated for higher voltages, because arcing between the powered surface and the grounded edge of the electrode appears.

The confinement voltage $U_{C}$ is applied to the inner segment of the lower electrode. The potential well for particle trapping can be manipulated by voltages of the order of 20 V that are applied by means of a decoupled dc voltage supply.

Both, the external bias voltage and the confinement voltage, are supposed to affect structural ordering of the dust particles, as well as influencing the damping of the ion two-stream
instability and the phonon stream instability, respectively. Therefore, the impact of the external voltages on the phase transition can play a crucial role, and voltage fluctuations can lead to strong statistical deviations in the results of the measurements.

5.2.1. Impact of the external bias voltage on phase transitions. The external bias potential can be used to change the vertical position of the whole dust cloud. The stability of vertical particle chains is affected by this voltage too. This way of particle manipulation has a global character, inasmuch as the plasma is changed as a whole.

To study the impact of the external bias voltage, the condensation transition is examined for different negative and also for positive voltages. Simultaneous CCD observations were used to keep the parameters of the dust cloud as constant as possible. Namely, the size of the cloud and the position of the scattering volume within the dust cloud were controlled.

Negative bias voltages between 0 and \(-40\) V were studied in a first measurement. The case of floating bias (self-bias) potential was also treated in this measurement. In self-bias conditions, the upper electrode is isolated for dc fields and cannot discharge to ground. The self-bias changed during the pressure variation from \(-25\) V at 5 Pa to \(-15\) V at 24 Pa. Positive bias voltages between 5 and 35 V were examined in a second experiment.

Figure 10 presents the thermal energy of the horizontal component of motion versus the discharge pressure. Figure 10(a) shows the negative bias voltages and self-bias conditions. The measurement by self-bias is denoted by ‘float’. In figure 10(b), the positive bias voltages are shown. The transition curves of the thermal energy for the vertical particle motion look very similar and are not shown here.

Figure 11 depicts the critical pressure obtained by applying the modified Boltzmann fit for different external bias voltages. Figure 11(a) refers to the measurement with negative bias voltages, and figure 11(b) is according to the positive voltages. In panel (a), the measurement with self-bias conditions is shown separately on the left side. The horizontal component (closed symbols) and the vertical component (open symbols) are shown.

Two ranges can be distinguished for negative external bias voltages. The critical pressure for condensation \(p_{\text{cond}}\) strongly varies for voltages between 0 and \(-20\) V. For voltages in the range of \(-20\) to \(-40\) V the critical pressure is about 15.5 Pa and does not change very much.

This is confirmed by CCD observations too. The dust cloud changes its vertical position and shape for low negative voltages, but for voltages above \(-20\) V, a stable regime is observed.

For positive external bias voltages, a stable region is found between 20 and 30 V. In this range \(p_{\text{cond}}\) is about 18 Pa and changes only a little. For higher voltages \(p_{\text{cond}}\) drops again.

The critical pressure is, in general, higher for the vertical component at all applied voltages, but both components show a similar behavior.

Thus, two stable regions are found that can be used in further measurement to ensure that the external bias voltage has a minor impact on the phase transitions.

5.2.2. Impact of the confinement voltage on phase transitions. The second important manipulation voltage, the confinement voltage, is studied in the same way. Figure 12 shows the transition curves of both components of motion for confinement voltages between 15 and \(-20\) V. The impact of the confinement voltage is negligible, as is apparent from the overlapping curves. Some irregularities appear in the vertical component due to dust acoustic waves, as will be discussed in the next section.
Figure 10. Condensation transition for negative (a) and positive (b) external bias voltages. The pressure was increased in discrete steps from 5 Pa to above 24 Pa. The thermal energy falls from about 50 eV to nearly room temperature. The condensation transition can be strongly affected by the external bias voltage.

The independence of the condensation transition from the confinement voltage is due to the local character of this manipulation voltage. No change in the sheath edge position or the sheath width is observed due to the variation of the confinement voltage. The global plasma parameters are nearly undisturbed.

On the basis of this analysis, we choose the experimental parameters in subsequent measurements to give the maximum of stability. The analysis also demonstrates the stability of the experimental conditions. The results are reproducible even after several melting and condensation transitions were performed.

5.3. Cooling effect near the lower electrode

The irregularities at pressures around 6 Pa in figure 12(b) are a phenomenon of the melted state and do not explicitly depend on the confinement voltage or the external bias voltage.
Figure 11. The critical pressure \( p_{\text{cond}} \) is plotted against the external bias voltage. Both parameters are obtained from the modified Boltzmann fit, which was applied to the transition curves in figure 10. A range of stable conditions is found for (a) negative and (b) positive external bias voltages.

This phenomenon appears if large-amplitude dust acoustic waves are present. Note that the thermal energy is measured over several cycles of the dust acoustic wave. Thus, it is not an effect of variations of the velocities in different phases of the wave, because these variations are averaged.

CCD observations show that the dust particles come very close to the lower electrode due to the oscillation into the vertical direction. A possible explanation for the drop of the thermal energy is the nonlinearity of the sheath in the vicinity of the electrode. Here, the particles find very different conditions and it cannot be assumed that the heating processes are the same. Interestingly, the effect on the horizontal motion is much lower. That speaks of a low coupling between both components of motion and different instability modes for the two components.

A more detailed study of the losses of energy near the lower electrode is carried out by vertical scanning through the dust cloud by DLS. The position of the scattering volume is varied gradually from the top of the dust cloud to the bottom. Figure 13 illustrates the result. The thermal energy is plotted as a function of the vertical coordinate for (a) the horizontal and (b) the vertical component. Three discharge pressures of 5, 10 and 12 Pa were analyzed. The particle system was in the melted state and dust acoustic waves were present.

The thermal energy in the horizontal direction (a) drops almost linearly with an increase of the height of the scattering volume for all three pressures. The thermal energy in the vertical direction (b), on the other hand, suddenly drops in the vicinity of the lower electrode. The position of the decline depends on the discharge pressure. For pressures of 10 and 12 Pa, the thermal energy starts to drop at about 3 mm above the lower electrode. For 5 Pa, the drop is already at 8 mm.

This indicates a source of cooling originating from the vicinity of the lower electrode. The range of this cooling is longer at low pressures; the vertical motion of dust particles positioned up to 8 mm above the lower electrode is affected at 5 Pa. The additional heat transport mechanism by the vertical particle motion due to dust acoustic waves is believed to play an important role in this cooling effect.
Figure 12. Condensation transition for confinement voltages between $-20$ and $15$ V applied to the segments in the lower electrode. The horizontal component (a) is undisturbed by the change in the confinement voltage. This is true also for the vertical component (b). A slight irregularity can be seen for the vertical component. This is due to energy losses in the vicinity of the lower electrode in combination with dust acoustic waves.

5.4. The particle number density and the height of the dust cloud

The particle number density $n_d$ is an experimental parameter that is hard to control. The injection of particles by a dust dispenser gives only a rough control over the amount of dust that is introduced into the plasma. In addition to the number density, the height of the particle cloud $h_c$ is assumed to have a strong impact on the condensation transition [10–12].

The impact of these parameters is studied in the following measurement. The size of the dust cloud was reduced after each condensation transition by pushing a part of the particles over the edge of the lower electrode. This was achieved by a short variation of the confinement voltage.

The height of the dust cloud and the number density were determined by the simultaneous CCD observation of a vertical plane. To calculate the number density, the average inter-particle
Figure 13. The thermal energy for different vertical positions in the dust cloud at pressures of 5, 10 and 12 Pa. The particle system was in the melted state and dust acoustic waves were present. For the horizontal component (a), the thermal energy increases nearly linearly toward the electrode. The vertical component (b) starts to decrease suddenly 3 mm in front of the electrode for 10 and 12 Pa. This decrease already starts at 8 mm for 5 Pa. The nonlinear sheath near the lower electrode affects the heating mechanisms and dust acoustic waves lead to additional heat transport.

separation $\Delta$ is determined by the analysis of the pair correlation function $g(r)$. Even though $\Delta$ is the average particle distance in the vertical plane, it is used to calculate a three-dimensional particle density via $n_d = ((4/3)\pi \Delta^3)^{-1}$. The CCD measurement was made in the ordered state, and $n_d$ is therefore only an estimate for the particle density during the phase transition. The particle density in the melted state has to be considered to be somewhat lower.

Figure 14 shows the thermal energy in the horizontal plane versus the discharge pressure for different particle densities $n_d$ and heights of the particle cloud $h_c$. The condensation transition shifts to lower pressures as the density is reduced from $n_d = 149$ to $94 \text{ mm}^3$ with a corresponding change of the dust cloud height from $h_c = 6.9$ to $3.5 \text{ mm}$.

The maximum energy, reached at low discharge pressures, decreases with the particle number density and the height of the cloud. The decay rate of energy toward the transition pressure is nearly the same for all conditions examined here.

The data in figure 14 are analyzed by the modified Boltzmann fit. In figure 15, the critical pressure versus the product of the particle number density $n_d$ and the height of the dust cloud $h_c$ is shown. The horizontal as well as vertical particle motion are analyzed.

The critical pressure $p_{\text{cond}}$ rises with an increase of the product $n_d h_c$. The increase is stronger at low values and a saturation is indicated at high values of $n_d h_c$. The same dependence is observed for the horizontal and the vertical particle movement. This result implies that if the dust cloud is sufficiently extended in the vertical direction and the number density is large enough, the impact of the particle density on the condensation transition is weak.

The role of $n_d$ and $h_c$ as observed in this experiment is consistent with the predictions of the theory by Joyce et al [10–12]. An increase of $n_d$ leads to a higher dust plasma frequency $\omega_d$, which is proportional to $\sqrt{n_d}$. This results in a decrease of the ratio of the dust–neutral collision rate and the dust plasma frequency $\nu_0/\omega_d$. To maintain the damping of the ion
Figure 14. Thermal energy of the horizontal component of movement versus the discharge pressure under the variation of the particle number density \( n_d \) and the height of the dust cloud \( h_c \). For lower values of \( h_c \) and \( n_d \), the condensation curves are shifted to lower pressures and the maximum of energy attained in the melted state is lower.

Figure 15. The critical pressure for condensation \( p_{\text{cond}} \) versus the product of the height of the dust cloud and the particle number density \( n_d h_c \). The critical pressure \( p_{\text{cond}} \) strongly drops for low values of \( n_d h_c \). The measurement indicates a saturation at values above 800 mm\(^{-2}\); the impact of \( n_d h_c \) is less important.

two-stream instability, the decrease of \( \nu_d / \omega_d \) has to be compensated for by a higher collision rate \( \nu_d \), i.e. a higher gas pressure is necessary. Thus, the critical pressure \( p_{\text{cond}} \) grows with \( n_d \).

Then again the boundary condition (right-hand side of (18)) itself depends on \( \omega_d \) and therefore on \( \sqrt{n_d} \), albeit it is a weaker dependence. An increase of \( n_d \) shifts the boundary to higher values. This also has to be compensated for by a higher collision rate \( \nu_d \), i.e. an even higher critical pressure \( p_{\text{cond}} \).

In dust simulations by Joyce *et al.* [10–12], it was shown that the ion two-stream instability is convective. Thus, the height of the dust cloud determines the critical pressure of condensation and the heating rate due to the instability. A larger height of the dust cloud is expected to shift...
the critical pressure $p_{\text{cond}}$ to higher values, considering that the dust ensemble is more sensitive to the ion two-stream instability. This is in agreement with the experimental observations in figure 15.

The experimental observations are in agreement with the theoretical description by Joyce et al [10, 11].

5.5. Discharge power

The discharge power is an elementary plasma parameter that has an effect on the density and the temperature of electrons and ions in the plasma. A variation of the discharge power has serious consequences for the structure and dynamics of the dust ensemble. To shed light on the impact of this parameter on the phase transition in a complex plasma, the condensation transition is studied under the variation of the discharge power in this subsection.

The discharge power was varied between 1 and 22 W. These values correspond to peak-to-peak voltages between 35 and 80 V$_{\text{pp}}$. After a change in power, the matching was adapted to the changed parameters. Two different particle diameters of 3 and 7 µm were examined in two independent experiments.

The other experimental parameters were kept the same except for the confinement voltage. The confinement voltage was 40 V for 3 µm particles and 30 V for 7 µm particles to ensure the most effective particle trapping. This difference should not affect the comparison of the results, because the impact of the confinement voltage was shown to be small. To ensure a minimal impact of the external bias voltage, it was chosen to be 19 V.

Figure 16(a) depicts the critical pressure for condensation $p_{\text{cond}}$ as obtained from the modified Boltzmann fit of the transition curve of $T_eV$. The horizontal and vertical components of motion are analyzed for both particle sizes. In panel (b), the product of height and particle number density $n_dh_c$ is plotted versus the discharge power. Again, the CCD technique was used to measure the height and particle density in the ordered phase state.

The critical pressure $p_{\text{cond}}$ shows a similar behavior for both sizes of particles. A rise in $p_{\text{cond}}$ is observed at low discharge powers, and $p_{\text{cond}}$ changes less and finally saturates for higher discharge powers. The curve of the 7 µm particles is shifted to lower values compared to the 3 µm particles. The vertical component condensates at slightly higher pressures than the horizontal component for both particle sizes.

A change in the discharge power involves a variation of the sheath width and sheath edge position. The major effect among others on the dust particle system is a change in the particle number density and the height of the dust cloud. As shown in figure 16(b), the product of density and height, in general, increases with the discharge power. For 3 µm (squares) particles, a nearly constant value of $n_dh_c$ is seen for low powers. Above 10 W a higher level of $n_dh_c$ is reached. For 7 µm (circles) particles, an increase is found that gets slightly weaker for higher powers.

The results shown in figure 15 imply that the condensation transition is only weakly affected for values of $n_dh_c$ above a certain threshold. This also comes through in the evolution of $p_{\text{cond}}$ in figure 16(a). For high powers, the product of $n_dh_c$ is large and $p_{\text{cond}}$ is nearly constant. This is true for both particle sizes. For the 7 µm particles, a similar increase for $p_{\text{cond}}$ and $n_dh_c$ is seen at low discharge powers. Here, the strong dependence of $p_{\text{cond}}$ on $n_dh_c$ for low densities and heights becomes obvious again. The condensation pressure $p_{\text{cond}}$ for 3 µm particles at low powers slowly drops similar to the quantity $n_dh_c$. The product of the density and height is still quite large, so $p_{\text{cond}}$ changes only a little.
Figure 16. (a) Impact of the discharge power on the critical pressure for condensation $p_{\text{cond}}$ for 3 µm particles (squares) and 7 µm particles (circles). The horizontal (closed symbols) and the vertical component (open symbols) of motion are shown. (b) The product of the particle density and the height of dust cloud $n_d h_c$ versus discharge power. To determine $n_d$ and $h_c$, simultaneous CCD measurements were made.

Note that the situation at 1 W looks quite different. From visual observations, it can be seen that the plasma distribution changes significantly below discharge powers of 2 W. The particle cloud is not as stable as it is for higher powers. So, one has to be careful in the interpretation of the measurement at 1 W.

The dependence of the condensation transition described in the simulations by Joyce et al [10–12] on the particle number density and height has already been discussed in the previous section. An increase of $p_{\text{cond}}$ for higher $n_d h_c$ is predicted. This is expected to be the main contribution to the increase observed in figure 16(a).

There are other parameters that have to be discussed regarding the theoretical predictions. Firstly, the ion density $n_i$ rises with the discharge power and it follows a higher ion plasma
The decay constant $c$ of the exponential decay of the thermal energy in the melted state. (a) The decay constant for 3 $\mu$m particles and (b) 7 $\mu$m particles. The horizontal component (closed symbols) and the vertical component (open symbols) of motion are shown.

frequency. After (18), the boundary gets shifted to lower values and the ratio $v_i/\omega_i$ reduces too. Both have to be countered by a higher neutral gas pressure to maintain the damping of the ion two-stream instability. Hence, the critical pressure is expected to rise with discharge power. This is another contribution to the growth observed in figure 16(a).

Secondly, the electron temperature $T_e$ has to be considered. Even though an increase of the discharge power rises mainly the electron density, the temperature is also affected. A larger $T_e$ leads to a higher particle charge ($Z_d \propto T_e$), and this increases the dust plasma frequency $\omega_d$. A reduced ratio $v_d/\omega_d$ and a shift of the stability boundary to higher values follow. Again, a higher critical pressure $p_{\text{cond}}$ follows. This contribution might be of minor importance due to the small change in $T_e$.

The role of $T_e$ can be more important for small discharge powers around 1 W. If the electron density becomes small, the electron temperature rises to sustain the plasma. This effect can be enhanced because of electron depletion by the dust component, which further increases $T_e$. As already mentioned, the situation at low discharge powers is expected to be complicated.

Finally, the ion streaming velocity has to be discussed. For higher discharge powers, the plasma potential relative to the electrode is higher. Thus, the ions gain more energy in the plasma-wall sheath, and the ion streaming velocity $u_i$ is higher. This increases the ion–neutral collision frequency $\nu_i$, which results in a more effective damping of the ion two-stream instability. Hence, the critical pressure decreases.

Additionally, the Mach number in (18) is larger for higher ion streaming velocities and this leads to a shift of the stability boundary to lower values. Altogether, an increase of $u_i$ leads to a reduced critical pressure $p_{\text{cond}}$. This is an opposite contribution to the other effects treated before, but in sum the observed increase can be explained by the theoretical predictions.

Figure 17 shows another fit parameter of the modified Boltzmann fit, the decay constant $c$ of the thermal energy. It describes the decrease of thermal energy when the discharge pressure is increased in the melted phase state. The decay constant $c$ is plotted versus the discharge power for (a) 3 $\mu$m particles and (b) 7 $\mu$m particles. In panel (b), the decay constant for powers below 6 W cannot be determined precisely due to statistical errors.
For both diameters, the decay constant is more negative for the horizontal component for all discharge powers. The difference between the components is nearly constant, in particular for the smaller particles. The more negative decay constant for the horizontal component represents a growing imbalance between the components when the pressure is increased. On the other hand, this means that in the highly energetic state the horizontal and vertical components are more equal than close to the phase transition, i.e. the components tend to be in a more isotropic state. This is observed for both particle sizes.

The decay constant levels off for high discharge powers and drops at low powers similar to the curve of $p_{\text{cond}}$.

Interestingly, the decay constant seems to be independent of the particle size and mass. For both particle sizes, the decay constant levels off at about $-0.13 \text{ Pa}^{-1}$ for the vertical component and $-0.23 \text{ Pa}^{-1}$ for the horizontal component. This is different compared to the observations at the critical pressure.

5.6. Anisotropy of the thermal energy

The anisotropy of the plasma-wall sheath leads to an anisotropy of the horizontal and vertical particle motion. The sheath electric field perpendicular to the electrode is responsible for enhanced heating in the vertical direction in the melted state. The thermal energy of the vertical component exceeds the energy in the horizontal plane by a factor of about 10. The energies are equalized in the ordered state, because the source of anomalous heating, the ion two-stream instability, is damped.

In this section, the ratio of the thermal energy in the horizontal and vertical directions $T_H/T_V$ is examined under the variation of plasma parameters.

Figure 18 illustrates the ratio $T_H/T_V$ during the condensation transition for the variation of the discharge power between 1 and 22 W. The plot reveals a remarkable feature of the anisotropy. The ratio $T_H/T_V$ is independent of the discharge power between pressures of 10 and 14 Pa, just before the condensing transition.

At low pressures a slightly higher spread can be observed, which can be explained by the cooling effect observed for the vertical component when the particles reach the vicinity of the lower electrode.

The condensation transitions can be seen at higher pressures. Here, the ratio of energies strongly rises and tends to equality. The thermal energy of vertical motion drops faster than the horizontal energy until they are equal.

This result is compared with other parameter variations that have been studied so far. A similar analysis to that illustrated in figure 18 is performed for the variation of the external bias voltage, the confinement voltage, the particle density, the particle size and the variation of the vertical position of the scattering volume. For all these parameters, the ratio $T_H/T_V$ is seen to be constant just below the critical pressure of condensation.

To compare all these results, an average curve of $T_H/T_V$ was calculated for the sequences of each parameter variation. Only the data points below the critical pressure of condensation were considered for averaging. So, the data of the power variation in figure 18, for example, are reduced to a single average curve.

Figure 19 depicts the averaged curves for all the parameters listed above. The power variation (figure 18) is represented by upside down triangles. The errors displayed here are calculated by Gaussian error propagation assuming an error for the thermal energy of 20% for the horizontal component and 10% for the vertical component (see [40] for details).
Figure 18. The ratio of the thermal energy in the horizontal and vertical directions $T_H/T_V$ during the condensation transition for discharge powers between 1 and 22 W is illustrated. The ratio is found to be independent of the power for pressures between 10 and 14 Pa. For higher pressures, the system tends to the ordered state with equalized thermal energies. At low pressures, a bigger spread of the data is caused by dust acoustic waves.

Figure 19. The average ratios $T_H/T_V$ of the sequences of each parameter variation. Only the points for pressure below the critical pressure of the phase transition are considered for the averages. The average ratios for the external bias voltage, confinement voltage, particle number density, discharge power and particle size and vertical position of the scattering volume are shown. The average ratios are independent (within error bars) of the experimental parameters studied.

The ratio $T_H/T_V$ shows nearly no dependence on any experimental parameter studied here. The calculated averages have overlapping error bars and are less than 0.04 apart for pressures between 8 and 19 Pa. The thermal energy in the vertical direction is between 10 and 25 times
larger than the horizontal energy in this range. The ratio $T_H / T_V$ generally rises for a decrease of discharge pressures. That is, the system gets more isotropic.

The independence of $T_H / T_V$ from various plasma parameters makes it a universal property of three-dimensional complex plasmas under gravity. In particular, the changes in the particle size, the vertical position of the measurement and the discharge power have no impact on the ratio $T_H / T_V$. These are important parameters that determine the structure and dynamics of the system.

These observations suggest that the amplitudes and growth rates of the ion two-stream instability modes in the horizontal and vertical directions have the same dependence on external plasma parameters. Furthermore, the heat transfer between the horizontal and vertical components of motion through dust–dust collisions has to be constant for all plasma parameters or has to be of minor importance. In the latter case, the ratio $T_H / T_V$ does not change significantly due to the coupling between the two components. To prove these assumptions and to get a deeper understanding of these observations, further simulations and subsequent experiments are necessary.

The constancy of $T_H / T_V$ is a remarkable property of the disordered state and has never been reported so far, whether in theory or in experiments. Examination of the vertical and horizontal components is difficult in most complex plasma experiments, and detailed analysis of the instability modes, which are responsible for the heating of each component, and the coupling between the two components is still an open issue.

5.7. Hysteresis loop in the phase transition

An important result of the simulations of instability-driven phase transitions is the prediction of a hysteresis loop [10–12]. The condensation transition is triggered by the ion two-stream instability and the melting is determined by the phonon stream instability. The damping of the two instabilities occurs at different discharge pressures, and therefore a hysteresis loop is observed.

For the experimental observation of the hysteresis loop, the discharge pressure was changed continuously from 16 to 26 Pa to cause a condensing transition, and after that, the pressure was decreased again to induce the melting transition. The data acquisition time of the whole measurement was 67 s, which is limited by the memory of the data acquisition hardware. The sample consists of $2 \times 10^8$ points measured with a frequency of 3 MHz. The horizontal and vertical components of motion were studied in two independent experiments.

The data sample of the continuous measurement is divided into several parts to calculate the autocorrelation functions. Each part consists of a window of $2 \times 10^6$ points. After the autocorrelation function is calculated, the window is shifted by $1 \times 10^6$ points and the next calculation is initiated. Thus, the parts are overlapping to increase the resolution of the analysis. This results in a temporal resolution of 0.33 s.

The DLS measurements presented in this section are made with scattering angles of about $7^\circ$. The discharge power was 2 W and the confinement voltage was 40 V. The external bias voltage was set to 20 V, which relates to the stable range discussed earlier.

Figure 20 shows the condensing and melting transitions for the horizontal (lower curves) and the vertical (upper curves) component of motion. The critical pressure of melting is lower compared with the critical pressure of condensation for both components of motion. Thus, in both experiments the predicted hysteresis loop was observed. The error of the thermal energy $T_{eV}$ was estimated to be $\pm 15\%$ (for details see [40]).
Figure 20. The condensation and melting transition induced by a continuous variation of the discharge pressure. The upper curves show the vertical and the lower curves show the horizontal component of motion. The critical pressure of condensation is higher than the critical pressure of melting for both components of movement. The error is assumed to be $\pm 15\%$. This hysteresis loop is predicted by simulations of Joyce et al \cite{10–12}.

The observed hysteresis loop is much smaller than the prediction by the simulations. The critical pressure of condensation for the horizontal component was determined to be $p_{\text{cond}} = 19.7$ Pa. The critical pressure for melting is $p_{\text{melt}} = 19.1$ Pa. The analysis for the vertical component gives $p_{\text{cond}} = 20.7$ Pa and $p_{\text{melt}} = 19.9$ Pa. Thus, the width of the hysteresis is only of the order of 0.8 Pa, but the data points are separated without overlapping error bars. The energies differ up to a factor of 3 near the phase transitions.

Only a close range around the critical pressures of the transitions is examined, due to the limitation of the sample size by the hardware. Hence, only the lower part of the dust cloud (around the scattering volume) was in the melted state at the start conditions of 16 Pa. The upper part of the dust cloud remained in the ordered phase state during the measurements. This was used to estimate the particle number density during the phase transitions applying the CCD method. The height of the dust cloud was estimated too. Both quantities were nearly constant during the measurements. The product of density and height $n_d h_c$ was of the order of 900. According to the earlier discussion, this indicates a reduced impact of density and height on the phase transitions.

A similar hysteresis loop is obtained for a pressure variation in the reversed direction, i.e. starting in the ordered state at high pressures. This shows that the observed effect is not caused by a delay in the data acquisition of the discharge pressure.

The width of the hysteresis loop can be manipulated by external plasma parameters. Although no systematic studies were performed on this topic, an increase of the discharge power seems to reduce the width of the loop. As mentioned, the height of the dust cloud plays a central role for the width of the hysteresis loop. A higher discharge power leads to the flattening of the dust cloud and this causes the hysteresis loop to vanish.
Figure 21. Map of the thermal energy in the horizontal plane for different positions of the scattering volume above the lower electrode. The pressure was varied from 25 to 5 Pa. The thick line (yellow) marks the melting transition.

5.8. Gradual melting in the vertical direction

In this section, gradual melting, a main feature of phase transitions in three-dimensional complex plasmas under gravity, is discussed. The transition does not appear simultaneously in the whole system, but a transition front moves vertically through the dust cloud. The melting transition, for example, is initiated at the very bottom of the dust cloud and the transition front moves upwards until the whole cloud is in the disordered state (see figure 4).

The aim of the measurements presented here is to analyze the movement of energy fronts through the particle system for the vertical as well as for the horizontal component of motion. It is not clear beforehand if the energy fronts move with the same speed for both components or if they move independently. Independent movement might be possible for different heating mechanisms in both directions, for example caused by independent instability modes.

In contrast to the previous measurements, which study the condensation transition of the dust ensemble, the melting transition is examined in this section. Two particle sizes of 3 and 7 µm are studied.

The confinement voltage was 40 V and the external bias voltage was 19 V, which lies in the stable regime. The pressure was decreased from 25 to 5 Pa for the 3 µm particles and from 20 to 4 Pa for the 7 µm particles in steps of 1 Pa. After a change of pressure, the system had 5 min to reach the new equilibrium. To realize the scan through the particle cloud, the whole discharge chamber was moved by a precise mechanical drive [40] in the vertical direction. This allows us to move the dust cloud relative to the scattering volume without disturbance of the particle system. The discharge chamber was moved downwards in steps of 0.5 ± 0.1 mm. One step is comparable with the diameter of the scattering volume.

The result of the scanning procedure is illustrated in figure 21. Here, the mapping of the thermal energy $T_{\text{eV}}$ for a melting transition of a dust ensemble consisting of 3 µm particles is presented in a contour plot. The ordinate denotes the vertical coordinate in the dust cloud and the abscissa is the discharge pressure. The thermal energy is color coded. This figure refers to the movement in the horizontal plane.
Figure 22. Slopes of the contour lines for energy values near the melting transition. Panel (a) displays the horizontal (squares) and vertical (circles) components of 3 µm particles and panel (b) shows the two components for 7 µm particles. A similar behavior is observed for both particle sizes. The energy transport is slightly faster for the horizontal component and nearly constant for all energies.

The thermal energy \( T_{eV} \) is low all over the dust cloud at high pressures. The system is in the uniform, ordered phase state. At intermediate pressures, the lower part of the dust cloud is melted with energies of the order of 10 eV, and the upper part is still in the ordered state. At low pressures, the thermal energy is distributed nearly uniformly again and the system is in the disordered state. The height of the dust cloud changes from 9 mm at 25 Pa to 11.5 mm at 5 Pa during the melting transition.

The contour lines are straight lines in the middle of the plot. This means that energy fronts move with constant progress from the bottom to the top. The melting transition is marked by a thick (yellow) line. It represents the energies at the melting pressure and is obtained from an analysis using the modified Boltzmann fit.

In a further analysis the contour lines are extracted and fitted by a linear function. Some of the contour lines need to be cropped at the bottom, because boundary effects lead to deviations from linearity. The slope of a contour line of a certain energy describes the distance the energy front moves upwards for a change in discharge pressure. In a totally uniform system, the melting would appear at all heights at the same time and the contour lines would be vertical.

The results of this procedure are discussed in two parts. Firstly, the lower energy range around the melting energies are treated, and secondly, the high-energy state is discussed.

Figure 22 depicts the slopes of the linear contour lines for low energies for (a) 3 µm particles and (b) for 7 µm particles. The horizontal (squares) and the vertical (circles) component of movement are plotted. The 3 µm particles in panel (a) have a slope of about 0.7 mm Pa\(^{-1}\) for the horizontal movement and about 0.6 mm Pa\(^{-1}\) for the vertical component at low energies below 0.5 eV. The curves have a slight increase of about 0.05 mm Pa\(^{-1}\) toward 3.0 eV. That means that higher energies are transported slightly faster through the dust cloud. A significant observation is the gap between the slopes for the horizontal and vertical components. The energy of movement in the horizontal component is transported faster by 0.1 mm Pa\(^{-1}\) compared to the vertical component.
Figure 23. Slopes of the contour lines for energy values above the melting transition. The horizontal and vertical components are shown for (a) 3\,\mu m particles and for (b) 7\,\mu m particles. For both particle sizes a faster movement of energy fronts is seen for the horizontal component (squares) and the movement gets faster for higher energies. On the other hand, the energy transport is nearly constant for the vertical component (circles).

It has to be noted that the two components are not necessarily in the same phase state at the same thermal energies. The energies corresponding to the critical pressure $T_{\text{melt}}$ are indicated in figure 22 by vertical lines. The line at 0.2 eV corresponds to the horizontal component and the one at 1.5 eV is related to the vertical component.

A very similar result is obtained from a measurement on 7\,\mu m particles, as illustrated in figure 22(b). The horizontal component has a slope of about 0.8 mm\,Pa\,^{-1}, where the vertical component has a slope of about 0.65 mm\,Pa\,^{-1} at low energies. The increase toward higher energies is a little stronger than that for the 3\,\mu m particles. The horizontal component again transports the energy faster than the vertical component. The difference between the slopes is about 0.15 mm\,Pa\,^{-1} and therefore slightly bigger than that for the 3\,\mu m particles. The horizontal transition energies are 6 eV for the horizontal component and 43 eV for the vertical component. The observed thermal energies for the 7\,\mu m particles are an order of magnitude larger than for the 3\,\mu m particles. These results indicate that the particle size and mass do not have a strong influence on the movement of the energy fronts through the dust cloud.

The different energy transport of the horizontal and vertical components can be explained by the different instability modes. As discussed, the ion two-stream instability is expected to be responsible for the anomalous heating in the melted state. As the authors have demonstrated in [10–12], not only a single mode can be present, but various modes can appear perpendicular and parallel to the ion flow. This can lead to different heating in the horizontal and vertical directions. The changes in plasma parameters such as Mach number, plasma density, etc can have different consequences for the growth rate of the instabilities. To prove this explanation, further simulations and experiments are necessary.

The second part of this analysis focuses on higher particle energies. Figure 23 illustrates the slopes of the contour lines above the melting energy. Again, the results for both components of movement are plotted for (a) 3\,\mu m particles and (b) 7\,\mu m particles.

The measurements yielded qualitatively the same results for the two particle sizes, again. But the horizontal component (squares) reveals a very different behavior than the vertical
component (circles), compared to the case of lower energies. The energy transport for the vertical movement is mainly constant for all energies, whereas the horizontal component reveals a strong increase of the slope of contour lines for higher energies. In the case of 7 \( \mu \)m particles, the slopes of the vertical component slightly increase for higher energies and the slopes of the horizontal component rise less compared to the 3 \( \mu \)m particles.

The coupling between the particles is low in the high energetic state (low discharge pressures), and the system acts as a non-ideal gas. The exchange of energy is enhanced by the free particle movement over long distances. Therefore, a more uniform system can be expected and the slopes of the contour lines should increase. This is observed for the energy transport of the horizontal component. In contrast, the slopes of the vertical particle motion stay mainly constant. The system becomes strongly anisotropic, with a horizontal component that shows enhanced energy transport and a suppressed energy transport for the vertical particle movement.

### 6. Conclusion

The DLS technique is applied to investigate the phase transitions of a three-dimensional complex plasma under gravity. The thermal energy of the dust component is used as the characteristic quantity to describe the phase transitions. The transitions are induced by the reduction of the discharge pressure and are studied under the variation of the most fundamental experimental parameters.

The thermal energy of the dust particles reveals a characteristic curve during the phase transition. A single-step transition is observed in a similar form for the melting as well as for the condensing transition. Below the critical pressure of the phase transition, the thermal energy increases exponentially with a decrease of the pressure.

The application of an external bias voltage to the upper electrode has an impact on the phase transitions. The critical pressure and the energy level that is reached in the melted state are affected. A stable range of voltages, in which the impact on the phase transition is small, is found for positive and negative voltages.

The applied confinement voltage has only a little impact on the phase transitions and can be chosen freely.

The particle number density and height of the dust cloud turn out to be critical parameters for the phase transition. The impact can be reduced by ensuring sufficiently large densities and heights.

A loss of energy of the vertical component of motion has been observed if the particles reach the vicinity of the lower electrode. The dust particles can come close to the lower electrode in the melted phase state since large-amplitude dust acoustic waves are present. The horizontal component of motion is not affected by this kind of energy dissipation.

As a main plasma parameter, the discharge power has been examined for two different particle sizes. The phase transitions show the same qualitative dependence on the discharge power for both particle sizes. The phase transitions of bigger dust particles are shifted to lower discharge pressures. The impact of the discharge power is mainly due to changes in the particle number density and the height of the dust cloud.

The theory of ion two-stream instabilities in a complex plasma by Joyce et al [10–12] makes predictions on how a change in the main plasma parameters influences the phase transitions in the dust system. The origin of the instabilities is the ion flow in the plasma-wall sheath. This leads to an additional heating of the dust particles due to the ion two-stream
instability. The instability can be damped by the combined effect of ion–dust and neutral–dust collisions which triggers the phase transition. The observations in the experiments are in good agreement with the predictions of the theory. Similar results in experiments with different particle sizes emphasize the fundamental character of the model.

A hysteresis loop in the phase transition of a complex plasma predicted by the model has been observed experimentally for the first time. Therefore, the assumption of different instabilities responsible for melting and condensation transition could be confirmed experimentally.

The gradual melting transition from the bottom to the top of the dust cloud is studied by scanning vertically through the dust cloud at different discharge pressures. The evolution of the energy distribution in the vertical direction is measured during the melting transition, and the movement of energy fronts in the vertical direction is examined. The energy fronts with energies comparable to the melting energy show nearly the same progress upwards for a change of pressure. Here, the energy fronts of the horizontal component move slightly faster than the energy fronts of the vertical component of movement.

At high particle energies, the movement of energy fronts of the horizontal component is much faster compared to the vertical component. Thus, the energy of the horizontal component of motion is distributed faster over the dust cloud than for the vertical component. The phenomenon of gradual melting in a ground-based complex plasma experiment has never been studied with this effort and accuracy previously.

The ratio of the thermal energy in the horizontal and vertical directions $T_H/T_V$ is found to be constant just below the critical pressure of the phase transition. This is observed for different variations of plasma parameters and seems to be a universal property of strongly coupled complex plasmas. The ratio $T_H/T_V$ increases for lower pressures, and the system tends to isotropy in the highly energetic state.

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