The application of dynamic light scattering to complex plasmas

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Abstract. The dynamic light scattering (DLS) technique is applied to the dust component of a complex (dusty) plasma, revealing a Gaussian intensity autocorrelation function for scattering angles between 4° and 175°. The Gaussian decay form represents free (ballistic) particle motion and allows determination of the one-dimensional squared particle velocity \(\langle v^2 \rangle \). At scattering angles below 1°, the intensity autocorrelation function is shown to be a combination of a Gaussian and an exponential function. This allows determination of the particle velocity and the diffusion constants at the same time. The dust system is fully described by the two components of motion in the horizontal and vertical directions. The two components are simultaneously measured on two scattering paths using only a single incident laser beam. In contrast to standard imaging techniques, the DLS method can be applied even to the disordered phase state where the dust particles have very high kinetic energies. In the ordered phase state, the assumptions of the DLS approach were verified by the independent Charge Coupled Device technique on the fundamental kinetic level. Furthermore, a careful discussion of the standard deviation of the DLS method proves that it can be used to study phase transitions of complex plasmas in detail.

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1. Introduction

Complex or dusty plasmas consist of electrons, ions, neutrals and macroscopic solid dust particles of nano- or micron size. The dust grains attain a charge in the plasma that can be positive or negative depending on the charging mechanisms. In most laboratory experiments, the dust is mainly charged by the inflow of electrons and ions, and acquires a negative charge typically of the order of $10^3$–$10^5$ elementary charges. The dust particle component is coupled much more strongly than other plasma components and the formation of ordered structures can be observed. These structures, also called ‘plasma crystals’, were discovered in 1994 by Chu et al [1], Hayashi et al [2] and Thomas et al [3].

In this work, micron size dust grains are confined in a low-temperature, radio frequency discharge. External forces that act on the dust grains, such as gravity and ion drag force, are compensated for by the electric force in the plasma-wall sheath of the lower electrode. Three-dimensional (3D) crystal structures of hundreds of layers were realized.

A fascinating feature of complex plasmas is phase transitions. The ordered structures can be melted by the variation of discharge pressure or discharge power. The dust grains attain unexpectedly high thermal energies that are about four orders of magnitude higher compared to the ordered state. Here, the anisotropy in the plasma-wall sheath plays an important role. The ion flow in the sheath leads to an ion two-stream instability [4–6] and a phonon instability [7, 8], which are responsible for the phase transitions and are leading to the enormous kinetic energy of the dust component in the disordered phase state.

The measurement of phase transition phenomena is quite demanding and thus only a few experiments have been performed until now, mainly on systems with only a few crystal layers [9, 10].

In contrast to other strongly coupled systems, complex plasmas can be studied directly on the kinetic level, because the micrometer grains can be seen with the naked eye. This is used in the Charge Coupled Device (CCD) observation technique [11]. Here, a laser beam is focused...
The standard CCD technique was used to obtain single shots of a vertical plane in the dust crystal. In (a), the system is in a well-ordered phase state revealing arrangement in vertical chains. In (b), the whole system is in the disordered state at low pressure and the dust grains show self-excited oscillations in the vertical direction.

on a sheet by a cylindrical lens system. This laser sheet is used to illuminate one layer of the dust system and the particle movement is detected by means of a CCD camera system that is mounted perpendicular to the laser sheet.

The video data are analyzed using video processing software and tracking routines [12]. The trajectories of individual particles are obtained and analyzed regarding the dynamics and structure of the 2D plane of the dust system.

Figure 1 illustrates CCD images taken from the vertical plane of a dust system in the ordered phase state (a) and in the melted phase state (b). The transition was induced by the reduction of the discharge pressure. The stable crystal is observed at sufficiently large discharge pressures. The anisotropic particle potential (wake field) in the plasma-wall sheath leads to the arrangement of the particles in vertical chains [13] as can be seen in figure 1(a) too. As shown in panel (b), the whole system is in a highly energetic disordered state at a lower pressure and the CCD camera system is not capable of following the fast particle motion. The particle positions can no longer be resolved.

To overcome the principal limitations of the imaging diagnostics regarding the temporal and spatial resolution, the ‘dynamic light scattering’ (DLS) technique is proposed in this work. DLS is well established in the other research fields of biology, chemistry and physics [14–16]. The particle system is illuminated by coherent laser light and the scattered light is detected by means of fast photon detectors. The movement of the particles along the scattering vector causes fluctuations in the scattered intensity. The temporal analysis of these fluctuations gives insight into the dynamics of the scatterers in one dimension. Depending on the ratio of the collision length of the scatterers and inverse absolute value of the scattering vector, a diffusive motion or a free motion is observed. In the first case, the diffusion constants can be measured, and in the latter case, the average particle velocity can be obtained.

This method was applied earlier on dusty plasmas in order to obtain diffusion constants and to determine the particle sizes of nano-scaled dust particles in [17]. Hurd et al [18] observed free particle motion for polydisperse dust particles with diameter below 200 nm and measured a Maxwell–Boltzmann velocity distribution.
Khodataev et al [19] measured diffusion constants in a thermal complex plasma that contains polydisperse dust grains with diameters of the order of 1 µm. Here, the system showed a comparatively strong coupling, related to a liquid phase state.

In this paper, the applicability of DLS to complex plasmas in a low-temperature, rf-discharge consisting of micron-sized, monodisperse dust grains is demonstrated. It is shown that the particle velocity can be measured in all phase states of the system. The error contributions of the measurement are carefully discussed. By a simultaneous CCD measurement in an ordered phase state, the DLS approach was benchmarked successfully. This verifies the assumptions of the DLS technique on the fundamental kinetic level.

This paper validates the DLS diagnostic to study complex plasmas in all phases from the crystalline state to the disordered state. Hence, the DLS approach can be used to study phase transitions of 3D complex plasmas in detail and to obtain a deeper understanding of the anisotropy of the dust system in the disordered state. This will be the subject of a subsequent paper.

2. Principles of dynamic light scattering (DLS)

Dynamic light scattering refers to methods where temporal fluctuations of scattered light are measured. The temporal evolution of the intensity of the scattered light is related to the motion of the scatterers.

The physical mechanism responsible for the fluctuations can be described as a Doppler effect. The Doppler shifts cause a broadened spectrum in the frequency domain which is equivalent to a fluctuating electric field in the time domain. This means that the time analysis of scattered light can be understood as a spectroscopic measurement in the limit of small line broadening.

The analysis of stochastic processes and fluctuating signals is the purpose of 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

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

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.

For an ensemble of particles, the scattered electric field results from the superposition of all scatterers

\[
E_S(q,t) := \sum_{j=0}^{N} a_j \exp(iq \cdot r_j(t)) \exp(-i\omega_i t). \tag{2}
\]

Here, \( N \) is the number of particles in the ensemble, \( q \) is the scattering vector, \( r_j \) is the position of the scatterers, \( a_j \) is the amplitude of the scattered electric field and \( \omega_i \) is the frequency of the incident light.
Figure 2. The impact of the absolute inverse scattering vector $q^{-1}$ on the result of a DLS experiment. The particle A is fixed and the second particle, B, experiences several collisions while it moves to the right. If $q^{-1}$ is large relative to the collision length (small scattering angle), the fluctuations will represent diffusive motion. On the other hand, if $q^{-1}$ is small, the movement along $q^{-1}$ is on linear trajectories and the measured fluctuations correspond to ballistic (free) motion.

The total scattered intensity $I(q, t)$ is the sum over the scattered intensities of all pairs of particles

$$I(q, t) := \sum_{i=1}^{N} \sum_{j=1}^{N} a_i a_j \exp \left[ i q \cdot (r_i(t) - r_j(t)) \right].$$

Equation (3) clearly demonstrates the connection between scattered intensity $I(q, t)$ and particle positions $r_i$ and $r_j$. The scattered intensity changes on the same time scales as the particle distribution, which is used in DLS to analyze the dynamics of the scatterers.

Two other important properties of the DLS technique are revealed by (3). Firstly, only particle movements along the scattering vector $q$ change the phase factor and therefore the scattered intensity. Hence, DLS is as per definition a 1D diagnostic. Secondly, the distance between a pair of particles has to change by a value of $q^{-1}$ to significantly change the phase factor in (3).

The inverse absolute value of the scattering vector $q^{-1}$ defines the length and time scale of the measurement. As a rule of thumb, a small $q^{-1}$ probes rapid motion on small length scales and a large $q^{-1}$ probes the long time regime and the large length scales. The length scale $q^{-1} = \lambda_0 / (4\pi n \sin(\theta/2))$ is a function of the scattering angle $\theta$, the wavelength of the incident light $\lambda_0$ and the refractive index $n$ of the background media. In this work $n = 1$ was assumed, because the refractive index of the low-temperature plasma used is very close to 1 for the wavelengths of interest. This is true for all the experimental conditions used in this work and the error introduced by this assumption is negligible.

To demonstrate the impact of $q^{-1}$, a simple situation with two particles is assumed as shown in figure 2. The particle A is fixed and a second particle, B, is moving to the right and suffers several collisions on its way. In the first case, a small scattering angle $\theta$ is chosen, which results in a large $q^{-1}$ (solid lines). The particle B has to move the distance $q^{-1}$ to cause a significant change in scattered intensity. The nature of this fluctuation is determined by the kind of motion...
taking place on this distance. Thus, the fluctuations will represent a diffusive particle motion. The autocorrelation function according to diffusive motion decays exponentially.

In the second case, the very same motion is probed with a large scattering angle \( \theta \), i.e. a small \( q^{-1} \) (dashed lines). The decay time of the fluctuations is of course smaller, and the type of fluctuation has changed because the type of motion on the distance \( q^{-1} \) is different. The trajectories along \( q^{-1} \) are linear now, i.e. the particle moves ballistically or freely. A Gaussian decay form is related to this kind of motion.

There are two major operation modes used in a DLS experiment. 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 that reaches the detector

\[
|E_D(q, \tau)|^2
\]

The time autocorrelation function of the detector signal

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

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 \( E_S(q, t) \)

\[
\Psi(q, t) := \sum_{j=0}^N \exp\left(i q \cdot r_j(t)\right).
\]

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

Under certain conditions, the homodyne and the heterodyne correlation function can be connected by a simple relation

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

This expression is known as the ‘Siegert relation’ [20]. It 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.

Most theoretical approaches for particle motion deal with the heterodyne autocorrelation function \( F_{\text{het}}(q, \tau) \). This means that if the Siegert relation cannot be applied, the connection of \( F_{\text{hom}}(q, \tau) \) with theory is rather difficult and a heterodyne experiment has to be preferred.

In a number of cases, 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. Even interacting particles can be treated like independent particles if the characteristic length scale \( q^{-1} \) defined by the scattering vector is small compared to the interaction length.

In these cases, the heterodyne scattering function \( F_{\text{het}}(q, \tau) \) simplifies to

\[
F_{\text{het}}(q, \tau) = \left\langle \sum_{i=1}^N \exp(i q \cdot [r_i(\tau) - r_i(0)]) \right\rangle
= \langle N \rangle F_S(q, \tau),
\]
where $\langle N \rangle$ is the average particle number in the scattering volume and $F_S(q, \tau)$ is called the self-intermediate scattering function.

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

$$g_{\text{hom}}(q, \tau) := \frac{\langle s(q, 0) s(q, \tau) \rangle}{\langle s(q) \rangle^2} - 1 = \beta_{\text{coh}}^2 F_S^2(q, \tau), \quad (10)$$

where $s(q)$ is the average of the scattered intensity and $\beta_{\text{coh}} (0 \leq \beta_{\text{coh}} \leq 1)$ expresses the finite degree of coherence at the detector. The spatial coherence of the scattered light is only given within a certain area, the so-called coherence area $A_{\text{coh}}$. The light beams from two points within the coherence area show the same fluctuations, whereas beams from points much further away are fluctuating statistically independent. Thus, if an area much larger than the coherence area is detected, the fluctuations are averaged [21]. The coherence area of a cylindrical scattering volume can be written as

$$A_{\text{coh}} = \frac{2 R^2 \lambda_0^2}{a (L \sin \theta + a |\cos \theta|)}, \quad (11)$$

where $\lambda_0$ is the wavelength of the incident light, $R$ is the distance between the detector and the scattering volume, $a$ is the diameter and $L$ is the length, respectively, of the cylindrical scattering volume. The ratio of the detector area to the coherence area has to be minimized by careful adjustment of the scattering arrangement.

The interpretation of $F_S(q, \tau)$ needs a theoretical description matching the physical properties of the system. In the case of free or ballistic particle motion, the particles move on linear trajectories along $q^{-1}$ with a velocity

$$v_j = \frac{r_j(t) - r_j(0)}{\tau}. \quad (12)$$

Thus, the self-intermediate scattering function can be written as

$$F_S(q, \tau) = \langle \exp(i q \cdot v_j \tau) \rangle. \quad (13)$$

This is the average of the expression $\exp(i q \cdot v_j \tau)$ over the velocity distribution $P(v)$. If a Maxwell–Boltzmann distribution is assumed, the heterodyne scattering function becomes

$$F_S(q, \tau) = \exp \left( -\frac{1}{2} q^2 \langle v_j^2 \rangle \tau^2 \right) \quad (14)$$

and the normalized autocorrelation function of a homodyne experiment is [14]

$$g_{\text{hom}}(q, \tau) = \beta_{\text{coh}}^2 \exp \left( -\frac{1}{2} q^2 \langle v_j^2 \rangle \tau^2 \right)^2 \equiv \beta_{\text{coh}}^2 \exp \left( -\frac{2}{\omega_q^2} \tau^2 \right), \quad (15)$$

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

The 1D average velocity-squared can be obtained from the width of the Gaussian $\omega_q$ via [14]

$$\langle v_j^2 \rangle = \frac{2}{\omega_q^2 q^2} = \frac{1}{2 \sigma^2 q^2}, \quad (16)$$

where in the last expression $2\sigma = \omega_q$ is used.
Figure 3. A sketch of the argon discharge and dust confinement. The dust particles are immersed by a vibrating dispenser through a hole in the upper electrode. The dust cloud is confined in the plasma-wall sheath. The horizontal confinement is realized by a potential applied to an inner segment of the lower electrode.

In the case of a large length scale $q^{-1}$ the self-intermediate scattering function reveals the diffusion constant $D$ [14]:

$$F_S(q, \tau) = \exp \left(-q^2 D \tau \right).$$

The heterodyne scattering function $F_{\text{het}}(q, \tau)$ is a single exponential function with a decay time $\tau_q = (q^2 D)^{-1}$. A homodyne measurement gives

$$g_{\text{hom}}(q, \tau) = \beta^2_{\text{coh}} \exp \left(-2q^2 D \tau \right) \equiv \beta^2_{\text{coh}} \exp \left(-\frac{\tau}{\tau_q} \right).$$

The diffusion constant $D$ in a homodyne experiment can be obtained from the decay time by

$$D = \frac{1}{2q^2 \tau_q}.$$  

3. Experimental setup

3.1. The complex plasma experiment

A capacitively coupled argon discharge is driven between two plane parallel metal plates as depicted in figure 3. To ignite and sustain the discharge, a radio frequency generator with a working frequency of 13.56 MHz is connected through a matching box to the upper electrode. The discharge power is between 1 and 20 W.

The dust particles are immersed in the plasma by a dust dispenser. This is 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 \( \mu \text{m} \). They have a monodisperse size distribution with a statistical deviation of only \( \pm 0.09 \, \mu \text{m} \).

The diameters of the upper electrode and the lower electrode are 120 and 140 mm, respectively. The electrode distance was optimized regarding dust confinement and optical access to be 54 mm.

The dust system can be observed through a top window and a hole in the upper electrode. This hole is covered with a grounded metal mesh to ensure a homogeneous plasma. The observation from the side is possible through four view ports with diameters of 100 mm.

The gas flow is introduced at the bottom of the vacuum system, far away from the main discharge chamber. This minimizes the turbulences in the chamber and the dust ensemble is not disturbed by a streaming background gas. The argon gas flow is between 2 and 5 sccm and the discharge pressure can be adjusted between 0.5 and 200 Pa.

The lower electrode is designed for particle confinement. The vertical confinement is realized by the electric field in the plasma-wall sheath. To realize the horizontal confinement, an inner segment of the lower electrode is set to a higher potential compared to the outer (grounded) ring. This forms a potential well for particle trapping. A voltage of up to 100 V can be applied to the inner segment by a decoupled voltage supply. 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 \( \mu \text{m} \).

### 3.2. The DLS experiment

The DLS setup as presented in figure 4 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 laser beam has a 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. This ensures identical scattering conditions for the components since in both the cases, the light with polarization perpendicular to the scattering plane is scattered. Furthermore, the scattering volume is exactly the same for the two components.

The single laser approach simplifies the DLS setup compared to the common approach regarding the number of mirrors, lenses and polarizers. This facilitates the arrangement and adjustment of the laser system and eliminates a significant source of error by omitting the second laser. A thorough analysis of the error contributions is shown in section 4.2.

The helium–neon laser has a wavelength of 632.8 nm and an output power of 21 mW. The laser beam is attenuated down to a few mW, to avoid heating of the dust ensemble by the laser radiation. At small scattering angles, a power of only 0.1 mW is necessary. 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. Thus, the lower photomultiplier detects the dust movement in the horizontal plane, because \( \mathbf{q}_1 \) is parallel to the electrode. The upper photomultiplier examines the vertical particle motion. As illustrated in figure 4, the scattering vector \( \mathbf{q}_2 \) is not perfectly perpendicular to the...
The quantities $q_1(2)$ and $\theta_1(2)$ denote the scattering vector and scattering angle of the lower (upper) light path.

electrode, but is tilted by a small angle. As long as the scattering angle $\theta_2$ is sufficiently small, the horizontal contribution to $q_2$ can be neglected and the vertical particle movement dominates the intensity fluctuations. This is a compromise to realize the single laser approach. The exact vertical motion can only be studied with a second laser beam. The scattering angles $\theta_1$ and $\theta_2$ are below $10^\circ$ in the experiments.

The scattered light is detected through collection systems that consist of an entrance aperture $a_1$, an interference filter, a Glan–Thompson polarizing prism and an exit aperture $a_2$. The design of the collection system is essential to maximize the coherence factor $\beta_{coh}$ on the one hand, and to obtain a sufficiently large scattering volume on the other hand. The coherence factor $\beta_{coh}$ in this experiment was of the order of 0.5. The cylindrical scattering volume, with a length of $l = 15$ mm and a diameter of $d_l = 0.7$ mm, contained some hundreds of dust particles.

The emission from the plasma and the background light is blocked by an interference filter with central wavelength of 632.8 nm and a bandwidth of 1 nm.

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 data streaming directly on a personal computer.

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Table 1. Factorial moments of unscattered (coherent) laser light. The photon statistic is calculated for a measurement of $4 \times 10^5$ samples. The factorial moments are identical within errors with the theoretical predictions for a Poisson distribution.

| Factorial moment | Experiment | Theory       |
|------------------|------------|--------------|
| $N^{(1)}$        | 1.000      | 1.000        |
| $N^{(2)}$        | 1.001      | 1.000 ± 0.005|
| $N^{(3)}$        | 1.005      | 1.000 ± 0.013|
| $N^{(4)}$        | 1.017      | 1.000 ± 0.032|
| $N^{(5)}$        | 1.035      | 1.000 ± 0.086|
| $N^{(6)}$        | 1.066      | 1.000 ± 0.251|

3.3. Validation of the DLS setup

The components of a DLS setup have to be chosen carefully regarding their contribution of random noise fluctuations to the detected signal.

The laser light source can cause noise due to intensity fluctuations or mode instabilities; the optical components can give rise to unwanted scattered light by dust or scratches on the surfaces. The photomultiplier contributes to noise by fluctuations of the gain or the supply voltage or due to the dark current.

To ensure that these unwanted contributions are negligible, the DLS setup is validated by a procedure described by Oliver \textit{et al} \cite{22}, p. 95. The unscattered, coherent laser light is detected by the photomultiplier and the signal is analyzed statistically according to the photon events. If contributions from the experiment are absent, the photon statistic should be Poisson. This is evaluated by the calculation of the factorial moments $N^{(r)}$ of the photon distribution

$$N^{(r)} = \frac{\langle n(n-1)\ldots(n-r+1)\rangle}{\langle n \rangle^r} = 1,$$

(20)

where $\langle \rangle$ denotes the average over all samples, $n$ is the number of photons in a sample and $r$ denotes the order of the moment.

The result is shown in table 1. A number of $2 \times 10^8$ data points were acquired with 10 MHz. All photons within a sample with the size of 50 $\mu$s (500 data points) are cumulated. This results in reduced data with $4 \times 10^5$ samples. These data are analyzed according to the photon statistics. The values of the experiment are close to the prediction for a Poisson statistic. The theoretical errors in the table are calculated following \cite{23}.

This measurement proves that the components of the setup described above introduce only a negligible distortion to the measurements.

The second part of this section justifies the application of the Siegert relation (8). This relation is based on the assumption that the complex amplitudes of the scattered light show a Gaussian distribution. Thus, only if this assumption holds the self-intermediate scattering function $F_S(q, \tau)$ in a homodyne DLS experiment can be obtained via the Siegert relation.

To prove this assumption, the distribution of scattered light ($\theta = 8^\circ$) is analyzed in a similar procedure to that described above. This time, a set of $2 \times 10^6$ samples with lengths of 10 $\mu$s are the basis of the photon statistics.
Table 2. Factorial moments of scattered (incoherent) laser light. The photon statistic is calculated for a measurement of $2 \times 10^6$ samples. The factorial moments are in good agreement with the theoretical predictions for the distribution with a coherence factor of $\beta_{coh} < 1$. This measurement justifies the application of the Siegert relation.

| Factorial moment | Experiment | Theory |
|------------------|------------|--------|
| Incoherent (scattered) | $N^{(1)}$ | 1.00 | 1.00 |
| Laser light | $N^{(2)}$ | 1.35 | $1.35 \pm 0.03$ |
| | $N^{(3)}$ | 2.34 | $2.30 \pm 0.16$ |
| | $N^{(4)}$ | 4.95 | $4.72 \pm 1.07$ |
| $2 \times 10^6$ samples | $N^{(5)}$ | 12.5 | $11.4 \pm 2.9$ |
| | $N^{(6)}$ | 36.4 | $31.3 \pm 94$ |

A perfect Gaussian statistic can only be expected for an experiment with a coherence factor $\beta_{coh}$ of unity. In a real experiment, the coherence factor is normally smaller than unity ($\beta_{coh} \leq 1$). The partial coherence leads to an amplitude distribution that is narrower compared to a pure Gaussian statistic. The Siegert relation can be applied if the factorial moments of the distribution are described by

$$N^{(r)} = (1 + \beta_{coh}) (1 + 2 \beta_{coh}) \ldots (1 + (r - 1) \beta_{coh}).$$

(21)

The factor $\beta_{coh}$ can be obtained from the second factorial moment, $N^{(2)} = (1 + \beta_{coh})$. The results of the experiment are shown in table 2. The second factorial moment shows that the coherence factor was only $\beta_{coh} = 0.35$ in this experiment.

The results of theory and experiment are identical within the errors. This important result justifies the homodyne DLS method to study complex plasmas with micron-sized dust particles.

4. Results

4.1. The application of DLS to measure ballistic dust particle motion

One of the most fundamental problems in the interpretation of DLS data is to find an adequate model for describing the decay of the autocorrelation function. In previous DLS experiments on dusty plasmas, a Gaussian decay [18] or an exponential decay was observed as in [19, 24]. But these experiments were performed on nano-scaled dust grains with relatively weak coupling between the dust particles. The particles studied in this work have diameters of several microns. The dust particle charge is much higher and the interactions between the particles and between particles and the electric field in the sheath of the plasma are much stronger.

To determine the form of the autocorrelation function for strongly coupled complex plasmas, a DLS experiment on $3 \mu m$ MF particles was performed. The dust system was in an ordered phase state at a discharge pressure of 20 Pa and a discharge power of 2 W (40 Vpp). The scattered light was detected in the homodyne mode with a scattering angle of $9^\circ$. The sample time was 20 s with an acquisition frequency of 10 MHz.

The normalized autocorrelation function $g_{hom} (q, t)$ obtained in this measurement is shown in figure 5 and explicitly reveals a Gaussian decay form. The residuals corresponding to the
Figure 5. The normalized autocorrelation function $g_{hom}(q, t)$ measured in the ordered phase state of a complex plasma. The scattering angle was $9^\circ$ and the data acquisition lasted 20 s. A Gaussian fit describes the data with high goodness. The residuals are below 0.3% as shown in the lower figure. A Gaussian decay form represents ballistic or free particle motion. The corresponding decay time is $\sigma = 0.9$ ms.

Gaussian fit are below 0.3%. This means that ballistic particle motion is observed, even though a strong coupling between the scatterers is present.

This might be a surprising result at first glance. But as discussed earlier, this is a question of the characteristic length scale of the DLS measurement. For the parameters of this measurement, the inverse scattering vector was $q^{-1} = 0.6 \mu$m. This is small compared to the average particle distance of about 400 $\mu$m. Thus, on length scales of $q^{-1}$ the particle movement is ballistic.

The decay time, given as the half-width of the Gaussian $\sigma = 0.5\omega$, is 0.9 ms. Using (16), the average velocity is $504 \mu$m s$^{-1}$. Hence, the decay times for the density fluctuations in a complex plasma are expected to be of the order of 1 ms in the ordered state and even faster for the melted state. This is about two orders of magnitude faster than what is seen in most DLS experiments on biological or chemical systems of scatterers in solutions [14].

The assumption of a Maxwell–Boltzmann velocity distribution in the derivation of the Gaussian decay form justifies the introduction of a particle temperature $T_d$. The thermal energy related to the dust movement in one dimension is defined as $T_{dV} := 0.5k_B T_d = 0.5m_d \langle v_x^2 \rangle$, where $k_B$ is the Boltzmann constant and $m_d$ is the mass of a dust particle that is given by the manufacturer of the dust powder.

To prove the assumption of ballistic particle motion for different scattering angles, a test suggested by Nossal et al [25] was applied. The self-intermediate scattering function $F_S(q, \tau)$ is a function of $Z = q\tau$ in the ballistic regime, i.e. a function of the scattering angle $\theta$ and time $t$. If $F_S(q, \tau)$ is measured for different scattering angles and plotted against $Z$, all curves should superimpose in the case of ballistic motion. Figure 6(a) illustrates a measurement of $F_S(q, \tau)$ versus time for scattering angles between $1^\circ$ and $9^\circ$. The time axis is scaled logarithmic for the sake of better distinctness of the data. The particle system was in an ordered phase state at 20 Pa, and the horizontal component of particle motion was detected. The decay time varies from 2 to 20 ms, as the scattering angle changes from $9^\circ$ to $1^\circ$.
Figure 6. Proof of ballistic particle motion for scattering angles between 1° and 9° in the ordered phase state at a discharge pressure of 20 Pa. (a) The self-intermediate scattering functions $F_S(q, \tau)$ at different scattering angles for particle movement in the horizontal plane. The decay time rises from 2 to 20 ms, while the scattering angle is decreased. (b) The scattering functions $F_S(q, \tau)$ superimpose if they are plotted over $Z = q \tau$, which proves ballistic particle motion. Small deviations appear due to the measurement error of the scattering angle $\theta$.

In (b), the same data are plotted against $Z = q \tau$, and as can be seen, the scattering functions $F_S(Z)$ lie on top of each other with good accuracy. Small deviations are due to the error in determination of the scattering angle.

A similar experiment was done at a discharge pressure of 2 Pa beyond the melting transition. The dust particle system shows no order anymore and the dust particle energy reaches up to 100 eV in the horizontal plane. The related decay times are $2 \times 10^{-5}$ s for a scattering angle of 9° and $1 \times 10^{-4}$ s at 1°. The self-intermediate scattering functions plotted as a function of $Z$ also superimpose in this case, within the measurement error of the scattering angle $\theta$ (data not shown).

DLS measurements in the vertical direction also reveal autocorrelation functions with Gaussian decay forms. This means that neither the electric field in the plasma sheath in the
Figure 7. The self-intermediate scattering function $F_S(q, \tau)$ for scattering angle below $4^\circ$ in a state close to the phase transition. Next to the Gaussian decay an additional exponential contribution, which represents diffusive motion, appears. A combination of Gaussian and exponential functions fits more precisely than a pure Gaussian function. The decay time changes from $\sigma_G = 0.11$ ms for the Gaussian fit to $\sigma_{G\text{Exp}} = 0.09$ ms for the combined fit.

direction of movement nor the large kinetic particle energy and related higher collision rates affect the kind of motion on the length scale of the DLS measurement $q^{-1}$.

Furthermore, DLS experiments with scattering angles from $4^\circ$ up to backward scattering at $175^\circ$ were seen to take place in the ballistic regime in all phase states. The choice of the scattering angle is a question of optimizing the intensity of the scattered light and the data acquisition rate of the hardware. Backward scattering at $175^\circ$ allows examinations on very fast time scales, because the length scale $q^{-1}$ is of the order of $0.05 \mu$m. This can be used to study the particle dynamics with a high temporal resolution of about 1 kHz. Note that only the horizontal movement can be studied in the backscattering mode with the setup presented in this work.

Small deviations from a pure Gaussian decay are noticed at scattering angles below $4^\circ$ if the system is close to the phase transition. Here, a second contribution to the autocorrelation function appears as shown in figure 7. The x-axis is scaled logarithmic to see the differences between the data and the fit more easily. The pure Gaussian function cannot fit the data with high precision. The Gaussian fit delivers a decay time of $\sigma_G = 0.11$ ms. A fit with a combination of Gaussian function and exponential decay gives better results. The decay time for the combined fit is $\sigma_{G\text{Exp}} = 0.09$ ms. Hence, even in these cases, the Gaussian decay time $\sigma$ can be extracted, but it is recommended to measure at larger scattering angles to reach the pure ballistic regime.

Note that as long as the two decay forms are separated in time and the fit procedure gives confident results for both decay forms, they can be treated separately and the assumptions made in the theory section of pure ballistic (the Maxwell–Boltzmann distribution of particle velocities) and diffusive motion, respectively, are not violated.

There are some other effects that can influence the autocorrelation function, but remain of minor importance for most cases. One example is given by fluctuations of the autocorrelation function around the baseline, as can be seen in figure 6. They are due to number fluctuations, dust density fluctuations on fast time scales or streaming dust particles. These fluctuations are important for short time sampling, where they can lead to large statistical errors.
The contributions to the error of a DLS experiment will be discussed in the next part in more detail.

The CCD diagnostic technique offers a unique way to benchmark the DLS method in the ballistic regime. The examination of the scatterers on the kinetic level is not possible in common DLS experiments on nanoparticles or macromolecules. The assumption made in the DLS theory and setup can now be verified directly by the independent CCD diagnostic for the first time.

In this experiment, the movement in the horizontal plane is studied. The number of particles probed is about 700 for DLS and about 900 for the CCD measurement. The CCD method attained information from a plane with a dimension of $10 \times 12 \text{ mm}^2$. The average particle distance in the horizontal plane was of the order of $400 \mu\text{m}$.

The particle motion in the horizontal plane is expected to be isotropic. This was justified by earlier CCD camera observations. Hence, the DLS velocities can be extended to 2D velocities and the related thermal energies can be compared to the CCD results. The results of this experiment are illustrated in figure 8(a). The discharge pressure was decreased from 55 to 1 Pa at a constant discharge power of 4 W, which forced a melting transition in the dust particle system. The 2D thermal energy of the particles is shown for the DLS (squares) and CCD technique (open circles).

At low pressures and fast particle movement, the CCD camera method fails to determine the particle velocity, and a saturation of the thermal energy is seen. The CCD approach is limited to the ordered phase state, due to the low frame rate of the camera. In the experiment here, a frame rate of $52 \text{ s}^{-1}$ was used, which is sufficient to resolve the motion in the strongly coupled state above 22 Pa.

The thermal energies measured by DLS in the pressure range below 15 Pa are much higher and indicate an exponential growth with a decrease in pressure. The fast particle movement causes no principal problems for a DLS experiment. Even at 2 Pa the autocorrelation is Gaussian, and thus, even in the melted state a Maxwell–Boltzmann velocity distribution is observed.

At pressures above 20 Pa, the particles arrange to an ordered structure. In this case, good agreement between DLS and CCD measurements is found as shown in figure 8(b). Both curves show a similar decrease and the data points are close together with overlapping error bars. This justifies the assumption of an isotropic system in the horizontal plane. The errors for the DLS approach used here are discussed in the following part. For the measurement shown, the error of DLS is $\pm 15\%$. The error for the CCD observation is mainly due to wrong particle identification and bad particle tracking through the image sequence. It is estimated to be $\pm 10\%$.

4.2. Statistical error of the DLS method

The errors in a DLS experiment are estimated by statistical methods and Gaussian error propagation. There are three dominant sources of errors in the determination of the thermal energy of dust particles $T_eV$ by means of DLS. Firstly, the limited number of dust particles in the scattering volume leads to a statistical fluctuation of the measured thermal energy. This can be countered by a longer data acquisition time $T_{DAQ}$ to improve the temporal average. This error contribution is described by statistics of the decay time $\sigma$ as obtained from the Gaussian fit of the autocorrelation function. The standard deviation of $\sigma$ is determined for different acquisition times $T_{DAQ}$. This analysis was repeated for three different scattering angles $\theta$. 

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Figure 8. Benchmark of the DLS method (squares) by simultaneous CCD observation (open circles). (a) The curve of the CCD measurement saturates in the melted state at low pressures. The determination of the correct velocities fails due to wrong particle tracking. The DLS measurement shows an exponential increase of the thermal energy with decreasing pressure. (b) The very good agreement found in the ordered state (higher pressures) justifies the DLS approach. Both curves decline in a similar way and the data points are close together with overlapping error bars.

Furthermore, the calculation of the thermal energy requires the measurement of experimental parameters. This leads to systematic errors. The second and third major sources of error are therefore given by the error in the determination of the scattering angle $\theta$ and the particle radius $a$. These contributions are considered in a Gaussian propagation of error calculation as described later.

To examine the statistical fluctuations of $T_{\text{ev}}$, a long measurement was divided into parts with equal length. The length of the samples was varied between 20 s, to examine the error of long acquisition times, and 1 ms, to get the error of short measurements. The average and standard deviation of the Gaussian decay time $\sigma$ were calculated for a set of data samples of the same length. The experiment was done for scattering angles of $4^\circ$, $9^\circ$ and $175^\circ$. The particle system was in the ordered phase state at a discharge pressure of 40 Pa for small angles and 20 Pa at backward scattering.
Table 3. Relative statistical errors of the decay time $\sigma$ for different acquisition times $T_{\text{DAQ}}$. The average $\chi^2$ value describes the goodness of the Gaussian fit analysis. The results for scattering angles of 4°, 9° and 175° are shown.

| $T_{\text{DAQ}}$ [s] | Relative error of $\sigma$ [\%] | $\chi^2$ [10^{-6}] | Relative error of $\sigma$ [\%] | $\chi^2$ [10^{-6}] | Relative error of $\sigma$ [\%] | $\chi^2$ [10^{-6}] |
|----------------------|----------------------------------|---------------------|----------------------------------|---------------------|----------------------------------|---------------------|
| 0.001                | –                                | –                   | –                                | –                   | 32.2                             | 107                  |
| 0.01                 | –                                | –                   | –                                | –                   | 33.0                             | 107                  |
| 0.2                  | 17.3                             | 80                  | 13.8                             | 19.2                | 7.6                              | 11                   |
| 0.5                  | 11.5                             | 31                  | 8.7                              | 7.00                | 3.7                              | 2.1                  |
| 1.0                  | 7.7                              | 16                  | 6.4                              | 3.72                | 2.7                              | 1.0                  |
| 2.0                  | 6.2                              | 9.2                 | 4.7                              | 2.13                | 2.2                              | 0.5                  |
| 5.0                  | 3.6                              | 4.5                 | 3.1                              | 1.00                | 1.5                              | 0.2                  |
| 10.0                 | 1.7                              | 3.5                 | 1.5                              | 0.43                | 1.2                              | 0.1                  |
| 20.0                 | 0.8                              | 3.3                 | 0.8                              | 0.34                | 0.6                              | 0.06                 |

The results of the statistics are shown in table 3. The relative error of $\sigma$ is the ratio of the standard deviation and the average of $\sigma$ given in percentage. The $\chi^2$-fit parameter describes the goodness of the Gaussian fit. For scattering angles of 4° and 9°, the decay of the autocorrelation function could not be resolved for acquisition times below 0.01 s. The relative errors are about 1% for 20 s of acquisition and 20% for short measurements of 0.2 s. To get a high temporal resolution, backward scattering can be used. At 175°, a relative error of about 30% has to be expected for an acquisition time of 1 ms.

The $\chi^2$-parameter exhibits a less confident fitting for shorter acquisition times. Considering that the autocorrelation function is an average itself, a shorter acquisition time results in a more noisy autocorrelation function. This leads to a larger standard deviation of $\sigma$.

The data in table 3 indicate that the ratio $T_{\text{DAQ}}/\sigma$ has to be sufficiently large to obtain a small relative error. Note that the decay time $\sigma$ drops when the scattering angle is increased, because $\sigma \propto q^{-1} = \lambda_0 / (4\pi n \sin \theta/2)$. The average decay times of the measurements were 2.2 ms at 4°, 0.9 ms at 9° and 0.08 ms at 175°.

The decay time $\sigma$ is smaller in the melted state, and therefore a smaller statistical error for the same acquisition time can be assumed. But additional contributions to the error can appear in the highly disordered state. The presence of dust acoustic waves (DAW) can lead to larger statistical errors if the acquisition time is shorter than the oscillation time of the wave. The acquisition time has to be of the order of one second to average this contribution to the error.

Furthermore, the large-amplitude oscillations can bring the dust particles to regions with different plasma conditions, e.g. different plasma-wall sheath conditions in the vicinity of the electrodes. This affects the heating of the dust particles, since the instabilities depend on the plasma parameters. This can affect the average particle temperature even if the acquisition time is much longer than the oscillation time [26].

Next to the statistical fluctuations due to the finite size of the particle ensemble, systematic errors have to be considered. The systematic error in the calculation of the 1D average velocity $\sqrt{\langle u_x^2 \rangle} = 1/(\sqrt{2}\sigma q)$ is mainly caused by the measurement error of the scattering angle $\theta$ in
Table 4. Relative error of the 1D particle velocity $v_x$ and the corresponding thermal energy $T_{eV}$ for different acquisition times $T_{DAQ}$. The errors are obtained by Gaussian propagation of error calculations on the basis of the data for $\sigma$ from table 3. The errors for scattering angles of $4^\circ$, $9^\circ$ and $175^\circ$ are presented.

| $T_{DAQ}$ [s] | Relative error $v_x$ [%] | Relative error $T_{eV}$ [%] | Relative error $v_x$ [%] | Relative error $T_{eV}$ [%] | Relative error $v_x$ [%] | Relative error $T_{eV}$ [%] |
|---------------|--------------------------|-----------------------------|--------------------------|-----------------------------|--------------------------|-----------------------------|
| 0.001         | –                        | –                           | –                        | –                           | –                        | 47.15                       |
| 0.01          | –                        | –                           | –                        | –                           | –                        | 67.2                        |
| 0.2           | 25.3                     | 37.5                        | 20.1                     | 29.9                        | 10.6                     | 17.2                        |
| 0.5           | 17.2                     | 26.9                        | 12.5                     | 19.9                        | 5.2                      | 11.1                        |
| 1             | 12.2                     | 20.7                        | 9.5                      | 16.2                        | 3.8                      | 9.9                         |
| 2             | 10.3                     | 18.5                        | 6.6                      | 13.0                        | 3.1                      | 9.5                         |
| 5             | 7.4                      | 15.5                        | 5.2                      | 11.6                        | 2.1                      | 8.9                         |
| 10            | 6.0                      | 14.2                        | 2.8                      | 9.9                         | 1.7                      | 8.7                         |
| 20            | 5.5                      | 13.8                        | 2.8                      | 9.9                         | 0.8                      | 8.5                         |

$q = 4\pi n/\lambda_0 \sin(\theta/2)$. The scattering angle was determined geometrically by measuring two sides of the triangle defined by the incident laser beam, the light path of the scattered light and the position of the first aperture. The errors in measuring the length of the triangle sides are assumed to be less than $\pm 5$ mm. The formalism of Gaussian propagation of error is used to obtain the error of the scattering angle $\delta \theta$ leading to the error of the scattering vector $\delta q$ that finally gives the error of the particle velocity $\delta v$. The error of the laser wavelength $\lambda_0$ was neglected in the calculation of $\delta q$.

To obtain the error of the thermal energy $T_{eV} = 0.5m_d \langle v_x^2 \rangle$, the error of the particle mass has to be considered. The manufacturer of the particles gives an error of the radius of $\pm 0.09 \mu m$ for the particles used. The particle density is $n_d = 1510 \text{ kg m}^{-3}$ with an error assumed to be less than $\pm 1\%$. All this is used in Gaussian propagation of error calculations again.

The results on the relative errors of the average particle velocity and average thermal energy are shown in table 4. The calculations are done with the data on $\sigma$ in table 3. The impact of the measurement error of the scattering angle $\theta$ can now be directly seen in the relative error of $v_x$, and further increase of the error of $T_{eV}$ can be related to the systematic error of the particle mass $m_d$.

The relative errors for the average velocity are between $3\%$ for 20 s and up to $25\%$ for 0.2 s at angles of $4^\circ$ and $9^\circ$. The error of $v_x$ at $175^\circ$ has not increased much because $q$ only depends weakly on $\theta$ near $180^\circ$.

The thermal energy has an error between $10\%$ for 20 s acquisition and up to $30\%$ for 0.2 s acquisition at angles of $4^\circ$ and $9^\circ$. For measurements with high temporal resolution at $175^\circ$ with acquisition times below 0.01 s, an error of the order of $70\%$ has to be considered. This error can be reduced significantly by using dust particles with smaller standard deviation of the particle radius.
4.3. Phase transitions

The thermal energy $T_{eV}$ of the dust particles during a melting transition, induced by a reduction of the discharge pressure, shows a characteristic form as demonstrated in figure 9 on a logarithmic scale. The discharge pressure was reduced from 24 to 5 Pa in steps of 1 Pa at a constant discharge power of 1 W. A DLS measurement with a scattering angle of 8°, an acquisition frequency of 10 MHz and a duration of 3 s was made at each pressure.

The thermal energy first starts rising slowly between 24 and 20 Pa. The vertical component shows a slightly stronger increase. A strong exponential increase of the thermal energy is seen for both components of motion at the critical pressure of the melting transition around 15 Pa. Below 13 Pa an exponential increase with a smaller growth rate of energy is observed. The thermal energy of the vertical component exceeds the horizontal energy by a factor of about 20 in the melted state. A single-step transition with the same characteristics is observed for the condensing transition too.

Simulations on a two-layer system show comparable thermal energies during the melting transition for the horizontal particle motion. But, in contrast to the measurements here, a two-step melting transition in the thermal energy is predicted in [8]. The first step is a growth of energy due to the phonon stream instability, where the structural order is still high. In the second step, the spatial order gets lost and a gaseous state is reached. The observations presented here indicate that the rise of energy due to the instability and the structural disorder occur at the same time.

Similar measurements can be used to study the phase transitions of 3D complex plasmas for different experimental conditions in detail. The examination of both components of motion at the same time gives a deeper understanding of the anisotropic energy distribution and the different growth rates of energy in the vertical and horizontal directions. These investigations will be addressed in a subsequent paper.
Figure 10. The logarithm of an autocorrelation function for a small-angle scattering experiment at $\theta = 0.83^\circ$. A mixed regime is observed. The ballistic regime appears as a Gaussian decay at short times and the diffusive regime becomes visible by an exponential contribution at longer times scales. The situation can be described by a combined fit of Gaussian and exponential decay with residuals typically lower than 0.3%. This results in decay constants of $\sigma = 16$ ms for the ballistic motion and $\tau_q = 59$ ms for diffusive motion.

4.4. Measurement of diffusion constants by DLS

A careful adjustment of the scattering setup makes a DLS experiment at $\theta = 0.83^\circ$ feasible. The related characteristic length scale is $q^{-1} = 7.0 \mu$m. The larger length scale $q^{-1}$ allowed to reduce the acquisition frequency down to 200 kHz. On the other hand, the sample time had to be increased to several minutes to obtain smooth autocorrelation functions.

An example of an autocorrelation function under these conditions is shown in figure 10. The data are plotted on a logarithmic scale to identify exponential decays more easily. Obviously, an exponential decay can be seen. A closer look shows a second decay form at short times that is not exponential. A combination of Gaussian and exponential fit is applied with good agreement. The residuals are below 0.3%.

This result shows that even for scattering angles at the lower limit, the pure diffusive regime cannot be reached, but there is still a contribution of ballistic scatterers. Nonetheless, the exponential decay is dominant and the diffusion constant can be determined.

As expected, the decay times are much smaller under these scattering angles. The exponential decay has a decay constant of $\tau_q = 59$ ms and the Gaussian part has a decay time of $\sigma = 16$ ms. With the equations $\langle v_x^2 \rangle = (2\sigma^2 q^2)^{-1}$ and $D = (2q^2 \tau_q)^{-1}$ the particle velocity and the diffusion constant are $v_x = 307 \mu$m s$^{-1}$ and $D = 4.1 \times 10^{-6}$ cm$^2$ s$^{-1}$.

To benchmark the small-angle DLS, the diffusion constants are compared to the values obtained by means of the standard CCD technique.

Figure 11 depicts the diffusion constants from the DLS technique (circles) and the CCD technique (open squares) for discharge pressures decreasing from 100 to 15 Pa. Good agreement is found; the data points show overlapping error bars. This proves the ability of the small-angle DLS approach to measure diffusion constants in a strongly coupled complex plasma.

There are two main sources of error to be considered for such a DLS measurement. The first contribution is due to the error in the determination of the scattering angle $\theta$. Again, the
Figure 11. The diffusion constants measured with the DLS approach (circles) and the CCD method (open squares). The pressure is varied from 100 to 15 Pa. The good agreement between the two techniques justifies the use of the DLS diagnostic to measure diffusion constants in a strongly coupled complex plasma.

The angle was determined by measuring distances in the triangle defined by the unscattered and scattered beams and the position of the first aperture. The accuracy of measuring distances is about ±5mm. The error of the decay time $\tau_q$ is assumed to be of the order of ±30%. The application of Gaussian error propagation to the equation $D = (2q^2\tau_q)^{-1}$ results in an error of the order of ±25% at pressures around 80 Pa and ±65% around 30 Pa. At low pressures, the Gaussian decay form becomes dominant and the fit of the exponential decay is less confident. This restricts the measurement of diffusion constants by DLS to the ordered phase state of the dust system.

The error can be reduced by a more precise determination of the scattering angle with alternative methods. Another way would be to choose a higher wavelength to reach the diffusive regime at larger scattering angles already. But DLS in the infrared or far-infrared requires a very different experimental approach regarding detectors, windows, light sources and so on.

5. Conclusion

The application of DLS on the dust particles of a complex plasma is demonstrated. The particle velocity in 1D can be determined in all phase states of the dust system.

The horizontal and vertical components of motion can fully describe the dust system. A single laser beam approach to determine the two components of motion at the same time was realized successfully. The setup was validated using photon statistics.

Furthermore, the assumption of Gaussian distributed complex amplitudes in the scattered light field was proven. This justifies the application of the Siegert relation to relate the homodyne scattering function to theory.

The decay form of the intensity autocorrelation function is found to be a pure Gaussian function for scattering angles between 4° and 175°. The Gaussian decay form represents the ballistic particle motion on the length scale of the DLS measurement $q^{-1}$. Ballistic particle motion is found for the ordered as well as for the melted phase state for the horizontal and vertical components of motion simultaneously.
The final benchmark of the method was done with aid of the well-established CCD technique in the ordered phase state. The agreement between both methods is excellent. This proves the assumptions made in the theory and the setup of DLS on the level of fundamental kinetics of single particle movement. Such a benchmark is not possible for common DLS experiments on nano scatterers in solutions [14].

The error in determining the particle velocity $v_x$ and the thermal energy $T_{eV}$, respectively, is estimated statistically and by Gaussian propagation of error calculations. For measurements of 2 s of data acquisition, the 1D velocity $v_x$ can be measured with an error of 10% at 4° and 3% in the backscattering mode at 175°.

The error of $T_{eV}$ is larger due to the uncertainties of the particle radius. An error between 10 and 20% has to be considered.

DLS offers a unique technique to study the highly energetic state of a complex plasma by examining the thermal energy. The thermal energy can be used as the characteristic quantity to analyze phase transitions in detail.

During a phase transition, the thermal energy $T_{eV}$ reveals characteristic features on a logarithmic scale. At low pressures a linear decrease of the thermal energy is observed. Close to the critical pressure a sudden decay down to energies that correspond to room temperature is seen. A single-step transition like this is observed for the melting as well as for the condensing transition.

The main difference between the transition of the horizontal and vertical components is an order of magnitude higher thermal energy for the vertical component in the disordered state.

Small-angle DLS experiments give access to a mixed regime between ballistic and diffusive motion. The diffusion constants and the thermal energy of the dust component can be determined simultaneously. The results are verified by the standard CCD technique in the ordered phase states.

The development of a theoretical model to fit the DLS data is considerably simplified by the fact that the particles used can be assumed to be identical. The small standard deviations of the average particle size and refractive index are considered in the determination of the statistical error in section 4.2.

In the case of polydisperse particles, i.e. particles with significantly different shape, size or refractive index, a more sophisticated analysis is required. The physical information of interest, e.g. the average particle velocity or the diffusion constant, cannot be obtained by a single fit of the autocorrelation function $g_{\text{hom}}(q, \tau)$ anymore. For example, the diffusion constant of plasma grown nanoparticles $D_c(r)$ has to be expected to be a continuous function of the particle radius $r$.

There are two common ways to analyze DLS data of polydisperse particles. In the case of only a few different particle sizes, $\ln \left[ g_{\text{hom}}(q, \tau) \right]$ can be expanded as a power series to obtain the average decay time $\langle \tau_q \rangle$ and therefore the average diffusion constant. This method also gives a measure of the dispersion of the average decay time. This approach is called the method of cumulants (see [14] for details).

The second and more powerful method applies an inverse Laplace transformation to $g_{\text{hom}}(q, \tau)$ to obtain the distribution of the decay time $\tau_q$. A mathematical approach to realize the Laplace transformation, known as regularization techniques, has been developed by Provencher (see [27, 28] for details).

This paper introduces an alternative diagnostic to the field of complex plasmas that overcomes the temporal and spatial limitations of the standard CCD technique. DLS is in
particular suited for the study of the fast particle movement in the disordered state and to obtain a deeper understanding of heating mechanisms and phase transitions in 3D dust systems. The coupling between the two components of motion and the transfer and growth of thermal energy in the disordered state are interesting topics for future research.

Moreover, the presented DLS approach allows a deeper study of the particle dynamics in DAW. The applicability of the experimental setup, as presented here, for a phase resolved study of DAW has been demonstrated in some preliminary experiments. The examination of both components of motion gives as insight into the coupling between the components in different phases of the DAW.

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