Topical Review

Controlling strongly correlated dust clusters with lasers

Hauke Thomsen1, Patrick Ludwig1, Michael Bonitz1,4, Jan Schablinski2, Dietmar Block2, André Schella3 and André Melzer3

1 Institut für Theoretische Physik und Astrophysik, Christian-Albrechts-Universität zu Kiel, 24098 Kiel, Germany
2 Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, 24098 Kiel, Germany
3 Institut für Physik, Ernst-Moritz-Arndt-Universität Greifswald, 17487 Greifswald, Germany
4 E-mail: thomsen@theo-physik.uni-kiel.de, ludwig@theo-physik.uni-kiel.de, bonitz@physik.uni-kiel.de, schablinski@physik.uni-kiel.de, block@physik.uni-kiel.de, schella@physik.uni-greifswald.de and melzer@physik.uni-greifswald.de

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Abstract
Lasers have been used extensively to manipulate matter in a controlled way – from single atoms and molecules up to macroscopic materials. They are particularly valuable for the analysis and control of mesoscopic systems such as few-particle clusters. Here we report on recent work on finite size complex (dusty) plasma systems. These are unusual types of clusters with a very strong inter-particle interaction so that, at room temperature, they are practically in their ground state. Lasers are employed as a tool to achieve excited states and phase transitions.

The most attractive feature of dusty plasmas is that they allow for a precise diagnostic with single-particle resolution. From such measurements, the structural properties of finite two-dimensional (2D) clusters and three-dimensional (3D) spherical crystals in nearly harmonic traps—so-called Yukawa balls—have been explored in great detail. Their structural features—the shell compositions and the order within the shells—have been investigated and good agreement to theoretical predictions was found. Open questions on the agenda are the excitation behaviour, the structural changes and phase transitions that occur at elevated temperature.

Here we report on recent experimental results where laser heating methods were further improved and applied to finite 2D and 3D dust clusters. Comparing to simulations, we demonstrate that laser heating indeed allows to increase the temperature in a controlled manner. For the analysis of thermodynamic properties and phase transitions in these finite systems, we present theoretical and experimental results on the basis of the instantaneous normal modes, pair distribution function and the recently introduced centre-two-particle correlation function.

Keywords: dusty plasmas, laser heating, phase transitions, strong correlations

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

1. Introduction

Complex (dusty) plasmas differ from traditional plasmas in a number of aspects: complex plasmas, in very general terms, are multicomponent plasmas that contain, in addition to electrons, ions and neutral atoms, also (macro-)molecules.

This may lead to substantial chemical reactivity or growth of clusters or nanoparticles that are of high interest for technological applications, see e.g. [1–4]. Alternatively, macroscopically large particles (dust) can be introduced into the plasma externally which may radically alter the plasma properties. These ‘dusty plasmas’ have evolved into a separate research field and are the subject of this review. Here, we
focus on non-reactive dusty plasmas containing comparatively large—typically micrometer sized—monodisperse plastic spheres.

The unusual properties of these plasmas arise from the behaviour of the dust particles and their enormously high charge. In the plasma, the particles are subject to continuous bombardment by the much lighter electrons, ions and neutrals. In a radio frequency (rf) discharge, the electron temperature is way above the ion temperature resulting in a higher impact rate of electrons onto the originally neutral particles, compared to the ions. As a consequence the particles become highly negatively charged, with the charge reaching values on the order of \( Q_d = O(-10,000 e) \) elementary charges for a particle of micron size \([5]\). It is due to this high negative charge that the dust particles are strongly interacting with each other, and the dust component of the plasma acquires a very large electrostatic energy that may exceed the thermal energy by orders of magnitude: the dust becomes strongly coupled (or strongly correlated), in striking contrast to usual high-temperature plasmas. At the same time electrons and ions are only weakly coupled and can often be regarded as a more or less uniform background.

Such strong correlation effects are presently of high interest in a large variety of fields, including condensed matter, dense plasmas (such as warm dense matter), ultracold quantum gases or the quark-gluon plasma. In fact, dusty plasmas serve as a prototype for studying correlation phenomena, e.g. \([6]\). A particular advantage of dusty plasmas is the large particle size and large inter-particle spacing (on the order of several micrometers to millimeters) which allows for a direct optical imaging of individual particles. At the same time the large mass results in slow characteristic time scales (on the order of hundreds of milliseconds), so the whole dynamics of these plasmas can be studied on the single-particle ‘atomic’ level \([1, 3]\). Despite the purely classical behaviour, dusty plasmas allow for unique and unprecedentedly accurate diagnostics of strong correlation effects which are important for other strongly correlated systems where such diagnostics are missing.

To have a quantitative measure of correlation effects in these highly non-ideal charged particle systems, it has been common to use the Coulomb coupling parameter \( \Gamma \) that relates the mean Coulomb interaction energy of two particles to the thermal energy as\(^5\)

\[
\Gamma = \frac{Q_d^2}{4\pi\varepsilon_0 b_{WS}k_B T_d},
\]

where the Coulomb interaction is estimated by the one of two particles separated by the Wigner–Seitz radius, that is related to the density by \( n^{-1} = 4\pi r b_{WS}^3/3 \) in three-dimensional (3D) \( (n^{-1} = \pi b_{WS}^2/2 \) in two-dimensional (2D)), and \( T_d \) is the kinetic temperature that corresponds to the dust particles’ random motion. In ideal (or weakly non-ideal), conventional plasmas \( \Gamma \ll 1 \). If \( \Gamma \) exceeds unity, the system is strongly coupled and the particle arrangement exhibits an increasingly long range order, giving rise to liquid-like behaviour. If \( \Gamma \) exceeds about one hundred the interaction is strong enough to spatially localize the particles leading to a crystalline structure. In macroscopic Coulomb systems, the critical value for the freezing transition is around \( \Gamma = 175 \) in 3D and \( \Gamma = 137 \) in 2D \([8–10]\).

Extended (nearly macroscopic) dust systems in 2D and 3D have been realized in experiments for decades, see e.g. \([11–16]\). There, \( \Gamma \) values of several hundreds are easily achieved, even at room temperature, as will be discussed in sections 5.2 and 6.

At these conditions the system is practically in its ground state. Therefore—and this may be surprising—for dusty plasmas, the major challenge for many applications is to lower the coupling strength. The main task is to do this in a controlled manner for instance by increasing only the dust temperature \( T_d \), without affecting the other plasma parameters. This is particularly important in order to study the thermodynamic properties, the melting process and the different phases of these systems. To achieve a controlled heating of dust clusters several methods have been used, including variation of the rf power or the neutral gas pressure \([17–20]\). However, these methods usually alter the whole discharge environment and, thus, effectively create a different plasma, making a ‘clean’ analysis of dust thermal effects difficult \([17, 21]\).

1.1. Laser heating of dust in the context of laser–matter interaction

A suitable approach to heat the dust without affecting the discharge environment is the use of lasers which is, therefore, in the focus of the present review. Since laser manipulation has become common in many fields it is of interest to put the present activities in the field of dusty plasmas into a broader perspective, before moving on.

The control of matter by lasers has seen dramatic progress over the last two decades which is due to the rapid increase of available coherent radiation sources. These are, in first place, optical and infrared lasers but, with the progress in the field of harmonic generation as well as of free electron lasers, also high photon energies—from UV to soft x-rays—became available. At the same time a tremendous variety of methods and mechanisms to control matter with lasers is being utilized which is briefly summarized in table 1. There we list the main properties of coherent electromagnetic radiation and how they are applied in different areas.

Traditionally, lasers have been used to excite electronic transitions in atoms and molecules allowing for high precision spectroscopy. By choosing a particular laser polarization, certain transitions can be activated or deactivated depending on the selection rules. In case of laser excitation of electrons into the continuum (photoionization) the photoelectron spectrum reveals detailed information on the target material. Related methods such as photoelectron spectroscopy are highly successful in atomic, molecular and condensed matter physics.
Presently being used in dusty plasmas are highlighted in bold, see also Table 2. Note that in many applications several laser properties are active simultaneously and the highlighted property should only be understood as the dominant one.

Table 1. Examples of laser–matter interaction processes in macroscopic systems (for details and references see main text). The concepts presently being used in dusty plasmas are highlighted in bold, see also table 2. Note that in many applications several laser properties are active simultaneously and the highlighted property should only be understood as the dominant one.

| Laser property       | Applications and mechanisms                              |
|----------------------|----------------------------------------------------------|
| photon energy $N\hbar \omega$, $N = 1, 2, \ldots$ | (multi-)photon excitation and ionization of atoms, molecules, condensed matter etc. |
|                      | photoemission spectroscopy                                |
|                      | inverse bremsstrahlung heating of matter                  |
|                      | photoionization of dust particles                         |
| laser polarization   | selection of intra-atomic (intra-molecular) transitions, e.g. linear vs. circular polarization |
| electrical field strength $E$, potential energy $-eEX$ | tunnel and field ionization of atoms                      |
|                      | field assisted creation of electron-positron pairs (Schwinger mechanism) |
| laser intensity, $I \sim E^2$ | heating, melting, evaporation of matter                  |
| Ohmic heat           | ionization of matter, creation of dense plasmas           |
| light pressure, $p \sim I$ | structuring of surfaces, micro-hole boring               |
|                      | acceleration of single dust particles                     |
| short pulse duration | time-resolved diagnostics (e.g. pump-probe schemes)      |
| field gradient of    | rapid excitation of non-equilibrium phenomena             |
| standing wave        | non-thermal melting of solids                            |
| field gradient of    | charge confinement, ‘optical tweezers’                    |
| moving wave          | confinement of neutral particles in optical lattices       |
|                      | charged particle trapping                                |
|                      | laser wake field acceleration                            |

With the increase of laser intensity also nonlinear processes such as multi-photon excitation and ionization have become available. In complex plasmas containing reactive species spectroscopic methods have found broad application as well, e.g. [2, 3] and references therein, but this has not been relevant for dusty plasmas so far. At the same time, ionization by UV radiation has been discussed by some groups [22–26], but has proven to be difficult. The interesting prediction is that the dust particles acquire a positive charge, but this will not be considered in the present paper.

Another important concept is the use of laser radiation with a spatially inhomogeneous intensity profile such as focused beams or interference patterns of several lasers. The latter gives rise to optical traps or optical lattices and has become a key tool in the field of ultracold atoms and molecules, e.g. [27] or in the field of colloidal systems, where laser patterns have been used to create quasi-crystals [28, 29]. Furthermore, the spatially varying field of a moving laser wave is at the heart of particle acceleration predicted long ago [30, 31]. In the mean time laser wake field acceleration has been successfully realized experimentally and allows for the generation of relativistic electron beams with MeV energy [32–36] as well as for ion acceleration. In dusty plasmas laser beams with spatially varying intensity profile are exploited utilizing the intensity dependence of the index of refraction of dust particles allowing to trap and move single particles (‘laser tweezers’) [37–39], see also [40].

An alternative direction in laser–matter interaction is heating of matter by generating Ohmic heat. Acceleration of electrons in solids easily allows to couple laser energy into the material and to melt it. This has become a standard method in technology and is used e.g. for microscopic structuring of surfaces as well as for drilling high quality micrometer-size holes. With the availability of ultrashort laser pulses it has become possible to couple the energy in a controlled way only into the electron subsystem, without heating the lattice, and to probe—on a femtosecond time scale—the ‘non-thermal melting’ of solids far from equilibrium, e.g. [41, 42]. Lasers can also be directly used to heat plasmas, via photon absorption in electron–ion collisions (inverse bremsstrahlung), e.g. [43–45] and references therein. With the help of Petawatt lasers, the combination of laser ionization and particle acceleration allows to produce and compress plasmas which reach densities that are comparable to and even exceed by one or two orders that of metals. This has led to the new fields of laser plasmas or high energy density physics, e.g. [46, 47]. It has been predicted long ago that sufficiently high laser intensities will allow to achieve even fusion conditions [48].

The heating of dusty plasmas follows the same general idea of transmitting momentum and energy from the radiation to the particle ensemble by exploiting the light pressure but, obviously, using very modest laser intensities. The specifics here lie in the large particle size, compared to the typical laser focus, and in the peculiar properties of finite dust clusters. These mesoscopic particle ensembles have many properties in common with more traditional neutral or metal clusters and will be discussed below.

1.2. Finite dust clusters and comparison with metal clusters

The properties of finite systems have been first studied in nuclear matter and more recently in the context of cluster physics. Finite 2D and spherical 3D dust clusters in a (nearly) isotropic harmonic trap provide a fascinating opportunity...
to systematically study the physics of mesoscopic few-particle systems. The reason is the above mentioned unique opportunity provided by dusty plasmas to study structure, dynamics and thermodynamics of all individual system particles on the kinetic level, i.e. resolving simultaneously all particle trajectories. This is of high interest for finite systems in a variety of other fields including gas or metal clusters, electrons in quantum dots, trapped ions, ultracold gases and so on. In none of these fields observations on the kinetic level are possible.

Finite 2D clusters have been studied first, already for quite some time [49–54]. More recently also spherical 3D dust crystals could be produced [55], and their low-temperature structure (which, in general, is more sensitive to screening than for 2D systems) is now well understood (for details see section 2.2). In the case of strong coupling, the excitation behaviour is dominated by collective modes of the whole system rather than by single particle dynamics. This is typical not only for finite dust clusters but for mesoscopic systems in trapping potentials in general. The frequency of some of these normal modes (in particular that of the monopole or ‘breathing’ oscillation) has been found sensitive to the internal properties as computer simulations, e.g. [73, 74]. There is overall consensus that the melting temperature of such clusters with size, geometry, and laser manipulation

| Property                      | Metal clusters | Dust clusters |
|-------------------------------|----------------|--------------|
| size, geometry                | $N \sim 10\ldots 10^4$ | $N \sim 10\ldots 10^4$ |
| elementary constituents       | single atoms   | spherical plastic particles |
| particle radius $R$           | $R \sim a_B$   | $R \sim 1\ldots 10 \mu m$ |
| interparticle distance $\bar{r}$ | $\bar{r} \sim \text{few } a_B$ | $\bar{r}$ several $100 \mu m$ |
| confinement                   | attraction from central ionic core | external potentials: trap, thermophoresis etc. |
| pair interaction              | Coulomb repulsion between electrons | screened Coulomb (Yukawa) repulsion |
| interaction strength          | fixed          | externally controlled |
| coupling parameter            | $r_s = \bar{r}/a_B \sim 2\ldots 5$ | dust component: |
| stationary state              | equilibrium    | non-equilibrium$^a$ |
| ground state                  | concentric shells | concentric shells |
| Laser manipulation            | $\sigma \gg R, \bar{r}$ | $\bar{r} \gtrsim \sigma > R$ |
| laser spot size $\sigma$      | homogeneous field across cluster | fast spot movement: |
|                               | • photoionization, | • single-particle diagnostics |
|                               | • collective ionization | by light scattering |
|                               | • Coulomb explosion | • particle control |
| laser heating                 | • via inv. bremsstrahlung | by optical tweezers |

by light pressure

Table 2. Comparison of atomic clusters (example of metal clusters) with finite dust clusters. In the lower part the relevant cluster manipulation approaches by means of lasers are outlined. Comments: $\alpha$–streaming electrons and ions may give rise to anisotropic and even attractive dust–dust interaction, depending on the plasma parameters and location in the discharge, e.g. [63–65].
also a decrease of the melting temperature was observed in simulations which is almost linear in the fraction of particles in the surface layer. For dust clusters, no such general trend is known yet due to the lack of systematic studies of larger clusters and due to the difficulties in controlled cluster heating. The laser heating technique described in this review will pave the way towards such studies.

When lasers where first shot on finite metal clusters, the experiments revealed a surprisingly effective energy absorption [76]. Subsequently, also collective absorption mechanisms (by coupling to plasmons) were observed [77, 78]. These effects could be successfully explained by theoretical modelling [79] and complex many-particle simulations, for an overview see [80]. Among the theoretical difficulties is the proper treatment of electronic quantum many-body effects, see e.g. [81, 82]. A detailed discussion of the various laser–cluster interaction mechanisms has been given in [83], see also the recent reviews [80, 84].

Among the most interesting effects of laser illumination of metal clusters is the emission of higher harmonics of the laser radiation [85], the emission of energetic electrons and, in case of strong ionization, Coulomb explosion of the whole cluster [86]. A similar rapid expansion of Coulomb and Yukawa dust clusters has been recently predicted in [87]. Here laser illumination is not necessary at all, it is sufficient to turn off the confinement potential. Similarly, many other effects observed for metal clusters in the presence of intense laser radiation can also be expected for dust clusters. However, more recently the focus was on a gentle heating of dust clusters so that the system is transferred from its ground state into a state with moderately elevated temperature.

As discussed above a focused low intensity laser beam is well capable to accelerate single dust particles. Similarly, stationary laser beams were used to excite shear flows in monolayer dust crystals [88, 89] or rotations in finite 2D clusters [53]. Mach cones could be excited by moving the laser spot through the dust crystal [90, 91]. A further application of moving laser spots is the realization of a heat source for the dust component [92–95]. A more detailed description of these pioneering laser experiments is given in section 3.1. An improved heating scheme allowing to realize a true thermodynamic heating with an isotropic, Maxwellian 2D velocity distribution has been applied by Schabliniski et al [96] to finite 2D dust clusters. A first heating concept for finite 3D clusters has been realized by Schella et al [97], see section 5.2.

The goal of this review article is to present an overview on these recent experimental developments, compare them to theory and computer simulations and to discuss possible future applications of laser manipulation of dust clusters. We start by giving a brief overview on the properties of finite 2D and 3D dust clusters in section 2. Then, we discuss the laser heating principle and how it is used to reduce the coupling strength in a controlled way (section 3). Dedicated numerical simulations of the laser heating are presented in section 4. Experimental results for finite 2D and 3D dust systems are presented in sections 5.2 and 6. We conclude in section 7 with an outline of future applications, including spatially inhomogeneous plasmas and time-dependent processes.

2. Structural, thermodynamic and transport properties of finite dust clusters

2.1. Structural properties of extended dust clusters

As described in the introduction, electrons and ions screen the repulsive interaction of the dust grains. The interaction between the dust particles is, in most cases, well described by a Yukawa-type pair-potential

$$\Phi_Y(r_i, r_j) = \frac{Q_i^2}{4\pi \varepsilon_0 |r_i - r_j|} \cdot e^{-\kappa |r_i - r_j|},$$

where \( \kappa = \lambda_D^{-1} \) is given by the inverse Debye length taking into account the screening effect of electrons and ions\(^7\). Wake effects due to the streaming ions (for recent overviews see e.g. [63–65]) are neglected in the presented results.

Extended 2D dust systems arrange, in the solid state, in a hexagonal lattice [11–13]. These systems are of special interest since the mechanism of thermodynamic phase transition in 2D systems from the ordered solid phase to the unordered liquid phase is still not finally clarified [98]. On the one hand, a first-order transition is predicted by the formation of grain boundaries between crystalline patches [98, 99]. On the other hand, a two-step second-order transition with an intermediate hexatic phase, the so-called KTHNY scenario [98, 100–102], is expected. A recent numerical study concludes that the hexatic phase is metastable and vanishes in the long-time limit [103]. Thermodynamic heating of 2D dust crystals by means of laser techniques might, therefore, be very beneficial in addressing these issues from the experimental side, see section 5.1.

2.2. Structural properties of finite dust clusters

The striking property of solid Yukawa clusters in 2D as well as in 3D is their well ordered structure. This structure and the loss of order with increasing temperature is accompanied by a sequence of phase transitions (or structural transitions) which are peculiar in finite systems [51, 104]. Details of these transitions are still open and of high interest for many finite size systems. They depend on the particular crystal structure which we, therefore, review in the following.

The Hamiltonian of the \( N \)-particle Yukawa cluster is (prior to laser manipulation) given by [3]

$$\mathcal{H} = \sum_{i=1}^{N} \frac{1}{2m} \vec{p}_i^2 + \frac{m\omega_0^2}{2} \vec{r}_i^2 + \sum_{i<j} \Phi_Y(r_i, r_j),$$

where the first term describes the kinetic energy, the second the confinement energy due to the harmonic trap of strength \( \omega_0 \) and the third the mutual Yukawa interaction energy. In this model, all dust grains are assumed to be equal in mass \( m \) and charge \( Q_i \).

Small 2D dust clusters typically consist of concentric rings [49, 51]. The core region of larger clusters with several...
hundred particles, in contrast, shows a hexagonal structure, like in infinite systems. This lattice has dislocations at the outer shells, where the circular boundary has to be matched [105].

Finite 3D dust clusters consist of spherical shells instead of rings [55, 106]. Due to their spherical shape, these clusters are called Yukawa balls (or Coulomb balls, when screening can be neglected). This structure has been measured experimentally [55, 107] and is reproduced in first-principle simulations–molecular dynamics (MD) or Monte Carlo (MC)– and has been investigated in detail for both Coulomb balls [108–110] and Yukawa balls [111, 112]. The general trend is that, when the screening parameter \( \kappa \) increases, more particles occupy the inner shells, and—even in the ground state—the average density becomes inhomogeneous, decaying towards the cluster boundary [113, 114]. This is in striking contrast to classical Coulomb clusters which have, at \( T = 0 \), a homogeneous mean density\(^8\).

Recently, also several analytical theories for the shell structure in 3D have been developed. The local density approximation as a continuum theory accurately describes the mean density profile of the spherical clusters [113, 114], but it misses the formation of shells. The positions and the populations of the shells of Coulomb balls are well reproduced by a slightly modified version of the hypernetted chain approximation which can be adapted to particles interacting via a Yukawa potential as well [115–118]. Beyond the radial shell structure, 3D dust balls exhibit a well ordered intra-shell structure at strong coupling. In contrast to a flat 2D system, the spherical curvature requires a fraction of pentagonal Voronoi cells in the hexagonal pattern on the shell [110, 119].

2.3. Thermodynamic properties and phase transitions

As the ground state properties of finite dust clusters are well understood by now [108–112], further investigations concentrate on thermodynamic properties at elevated temperatures. When a cluster is excited by feeding thermal energy into the system, metastable states with energies above the ground state energy \( E_0(N, \kappa) \) are occupied. These metastable states may differ from the ground state with respect to the occupation numbers of the shells or in the particle configuration within the shells. The metastable states of Yukawa balls as well as their increased population with temperature were investigated in both experiment [107, 120] and first principle simulations [121, 122]. Among others, these simulations allowed to determine the heights of energy barriers between different metastable states [121–123]. This ‘fine structure’ [110] is observed for 3D but not for 2D clusters.

When the temperature is increased a transition from a well ordered structure with thin shells and a highly symmetric intra-shell order (3D) towards a disordered particle fluid-like state is observed. It is an interesting question whether this process occurs rapid and constitutes a phase transition, as in macroscopic systems. While many similarities to phase transitions have indeed been observed\(^9\), there are also differences: the first is that in finite systems ‘melting’ requires a finite temperature interval. The second is that melting may involve a sequence of distinct processes. Therefore, in order to resolve and understand these processes, the concepts for characterization of phase transitions known from macroscopic systems have to be re-considered and adapted. In particular, structural parameters are required that are suitable to characterize the phase transition. The first quantity that comes to mind is the heat capacity, (the amount of heat \( \delta Q \) required to heat up the system a temperature \( \delta T \)) which is a widely used melting parameter e.g. in solid state physics. However, measuring the heat capacity is challenging in dusty plasmas. The dust subsystem exchanges energy with electrons, ions and neutrals making it very difficult to extract the pure heat capacity of the dust system with a particle number that is negligible compared to the number of the surrounding plasma constituents [75]. In contrast, the pair distribution function and, even more, the C2P correlation, discussed in the next section, have proven to be well suited for this purpose in theory as well as in experiments.

Let us now summarize the known results about ‘phase transitions’ in finite dust systems. In two dimensions, for the loss of the ring structure, two different melting processes were identified. The first process is attributed to the rotation of one ring with respect to the other rings [51–53]. The required energy for such a rotation crucially depends on the exact occupation number of the rings. For example, the commensurate configuration (‘magic number’) \( N = 19 \) (1-6-12) is very stable against such inter-shell rotations (due to the matching particle numbers on the inner and outer shell this cluster has a perfect hexagonal symmetry). In contrast, the \( N = 20 \) (1-7-12) configuration is extremely unstable against this excitation [53] and has a drastically reduced inter-shell rotation barrier and melting temperature. The second melting process is associated with particles undergoing a transition between two adjacent rings and is called ‘radial melting’. It, typically, takes place at substantially higher temperatures [21]. It is worth noting that the same kind of two-stage melting process is observed in finite quantum clusters [124, 125], indicating that these are correlation effects which are of high interest also beyond the field of dusty plasmas.

The complexity of the melting process increases when advancing from 2D to 3D clusters [123, 126]. Besides the melting of the radial structure and the inter-shell rotation, a third melting process emerges that is connected to the intra-shell order [126]. However, this classification is not strict since the interplay between the different melting processes in 3D clusters is utmost complex and there is, in general, no separation of the different processes. Thus many interesting questions are still open that have to be answered by experiments and theory.

In order to trigger phase transitions in dusty plasmas, selective control over the dust kinetic temperature

\(^8\) Nevertheless, as a reasonable first approximation for the average density one can often use the particle number \( N \) and the radius of the outermost shell \( R_C \), \( \langle n \rangle \approx N/(4\pi R_C^3/3) \)

\(^9\) As the solid and fluid ‘phases’ are, strictly speaking, concepts defined in the macroscopic limit the same holds true with phase transitions. Nevertheless, the processes in many finite systems are very similar and the application of thermodynamic concepts is useful. Often though the term ‘crossover’ is preferred in comparison to phase transition.
is essential. In particular, it is desirable to feed energy into the random dust motion without changing other plasma parameters such as the neutral gas pressure, the electron and ion temperatures and the flow velocity of the ions. A further requirement is that the entire cluster should be heated homogeneously while preserving an isotropic velocity distribution. As will be described in detail in section 3, this selective control over the dust temperature is indeed possible—by the means of intense laser light.

2.4. Key quantities for the analysis of finite dust clusters

An important structural parameter that characterizes the order (and its loss during melting) in extended systems is the radial pair distribution function \( g(r_{ij}) = g(|\vec{r}_i - \vec{r}_j|) \). It is commonly defined by the average number of particle pairs found at a distance of \( r_{ij} \) divided by the number of pairs which one would find in a homogeneous (i.e. uncorrelated) system with the same density. The first maximum of \( g \) reflects the mean inter-particle distance between the particles. An algebraic decay of the envelope of \( g \) over \( r_{ij} \) indicates a long range order, characteristic of the solid regime. Moreover, the height of the first maximum allows to detect a melting line in the \((\Gamma, \kappa)\) space [7].

The radial pair distribution function for finite 2D and 3D clusters is shown in parts d of figures 2 and 3, respectively. \( g(r) \) drops to zero after a few nearest neighbour distances for finite clusters in both 2D and 3D. For obvious reason, a true 'long range behaviour' cannot be investigated for finite clusters. Nevertheless, \( g(r) \) contains detailed information about the thermodynamic state of the cluster and its temperature dependence. In particular, a fine structure of the peaks in \( g(r_{ij}) \) is visible at high coupling strength (low temperature), indicating a frozen structure. When the temperature is increased, the subpeaks disappear. Finally, at a moderate coupling strength, see curve \( \Gamma = 10 \) in figure 2, the pair distribution function has only a single peak followed by a monotonous decay. The classification of inter-shell and intra-shell melting is not possible by means of this quantity as sampling the modulus of the distance does no distinguish whether the particles of a pair are on one shell or on different shells.

For this reason, the C2P correlation function \( g_2(r_1, r_{11}, \varphi) \) is introduced which takes into account the radial position of both particles as well as their angular distance with respect to the trap centre [97, 127]. A sketch of the sampled coordinates is shown in figure 1 for a 3D Yukawa ball consisting of two spherical shells and one particle in the centre. To evaluate the C2P, the sampled two-particle probability density \( \rho_2(r_1, r_{11}, \varphi) \) is computed and normalized by the uncorrelated probability density \( \rho_2^\text{uncorr}(r_1, r_{11}, \varphi) \).

\[
g_2(r_1, r_{11}, \varphi) = \frac{\rho_2(r_1, r_{11}, \varphi)}{\rho_2^\text{uncorr}(r_1, r_{11}, \varphi)}. \tag{4}
\]

The pair distribution function of finite clusters is computed by introducing discrete bins of width \( \Delta r \), counting the average number of particle pairs with a distance in the interval \([r_{ij}, r_{ij} + \Delta r]\) and dividing by the bin volume.

Figure 1. Yukawa ball with \( N = 60 \) particles consisting of two spherical shells and a single particle in the centre. In order to sample the centre-two-particle (C2P) correlation function based on the coordinates, for each particle pair, both radial coordinates \( r_1 \) and \( r_{11} \) as well as the angular pair distance \( \varphi \) with respect to the trap centre are recorded. The Voronoi grid of the intra-shell particle configuration is shown in grey.

Since \( \rho_2(r_1, r_{11}, \varphi) \) is invariant under rotation of the entire cluster, it is not necessary to remove a rigid rotation before sampling.

The C2P contains very detailed information on the structure of finite clusters and useful special cases. For example, integration of \( g_2 \) over both radial coordinates over a range corresponding to one shell allows one to extract the angular pair correlation function within that shell. On the other hand, when only one radius coordinate is integrated of the width of one shell, the C2P contains information on the relative radial and angular arrangement of the particles of the same shell as well as from different shells. To visualize this, \( g_2(r_1, \varphi) \) can be plotted in a color map, as is done for a 2D cluster, in figure 2, and a 3D cluster, see figure 3. In this color map, the intra-shell structure is responsible for the maxima and minima in \( \varphi \)-direction. While the data in the figures are from MC simulations, equally well one can use data from experiments with finite dust clusters, as is shown below in figure 12.

For the 2D cluster with \( N = 25 \) particles, depicted in figure 2, a reference particle from the inner shell is chosen by the integration range around \( r_1 \approx 0.7 \). Since the inner shell consists of three particles, the intra-shell neighbours appear as a peak at \( \varphi = 120^\circ \). At \( \Gamma = 1000 \), the distinct peaks found around \( r_{11} \approx 1.6 \) show that the angular orientation of the second shell with nine particles is locked with respect to the inner shell. This inter-shell order disappears between \( \Gamma = 1000 \) and \( \Gamma = 100 \) where \( g_2 \) is smeared out in angular direction. At moderate coupling, \( \Gamma = 10 \), hardly any angular correlations remain and also the radial order is lost. Particle transitions between different shells are revealed by a finite density in the radial regions between the shells.\(^1\)

Consider now a 3D Yukawa cluster with \( N = 60 \) particles, see figure 3. In the ground state (and at strong coupling), the particles are found in a configuration which has one particle in the centre, 15 particles on the inner shell and 44 particles...

\(^1\) Note that, at very strong coupling, the correlation function in the region between two shells cannot be calculated since both \( \rho_2 \) and \( \rho_2^\text{uncorr} \) become zero.
Figure 2. (a)–(c) C2P correlation function for a 2D Coulomb cluster with $N = 25$ particles for different coupling strengths. The ground state configuration consists of three concentric rings, see inset in (d). The first radial coordinate $r_I$ is integrated over a range corresponding to the inner shell indicated by the arrows. Intra-shell correlations are visible at $r_{II} \approx 0.7$ and $\phi = 120$. Pronounced inter-shell correlations with particles on the second shell are found at $r_I \approx 1.6$ for strong coupling. (d) The radial pair distribution function $g(r_{ij})$ shows distinct peaks at high $\Gamma$ which vanish when $\Gamma$ is being decreased. Note that $g(r_{ij})$ decreases to zero at large distances for all couplings, due to the finite size of the cluster. Results are from a MC simulation, $\Gamma$ is defined with the length unit $r_0 = [Q/(4\pi\epsilon_0 m\omega_0)]^{1/3}$ as the characteristic pair distance.

2.5. Transport properties

Besides thermodynamic properties, also transport coefficients and their dependence on parameters like temperature or the magnetic field strength are important characteristics of dusty plasmas. A particularly important example of transport coefficients is the diffusion coefficient. In dusty plasmas, diffusion was investigated in detail in macroscopic systems, e.g. [128, 129] and references therein. In particular, in two dimensions an anomalous diffusion was found [130–133] which turned out to be a transient effect [134]. Furthermore simulations were performed for magnetized dusty plasmas. Here, the diffusion coefficient $D_\perp$ perpendicular to the magnetic field as well as the parallel diffusion coefficient $D_\parallel$ were found to be strongly affected by the magnetic field in the strong coupling regime approaching Bohmian diffusion (decay with $B^{-1}$) [129]. Recently diffusion in a 2D one- and two-component magnetized strongly coupled plasma was studied and interesting behaviour of the diffusion coefficients of both components was reported [135].

One way to compute (or measure) the diffusion coefficient in a macroscopic system is to use the mean square displacement (MSD)

$$u_r(t) = \left\langle |\vec{r}(t) - \vec{r}(t_0)|^2 \right\rangle_N = 2 \text{dim} D t^\alpha,$$

as an average over all $N$ particles. Here dim is the system dimensionality (2 or 3) and $\alpha$ is the diffusion exponent which equals one for normal diffusion. However, the long time behaviour of the MSD has only limited meaning for mesoscopic systems since there the particles reach the cluster border after a few inter-particle distances [136].

Therefore, a different method is required to calculate the diffusion coefficients in small clusters. The instantaneous normal mode (INM) analysis has proven successful to this purpose [137–142]. This method deduces dynamic properties of a liquid state from the curvature of the energy landscape of the momentary configuration of the cluster. The first step is to calculate the dynamical (Hessian) matrix of the potential energy in the Hamiltonian, equation (3), as

$$A = \left( \frac{\partial^2 E}{\partial r_{ij} \partial r_{ij}} \right),$$

where $i, j$ are particle indices and $\alpha, \beta$ indicate the (two or three) components of the coordinates. The eigenvalues of the this matrix present the squared eigenfrequencies $\omega_\alpha^2$ of the system. In a stable state, all these eigenvalues are
positive resulting in real eigenfrequencies which reflect stable oscillations of the particles in the potential cage formed by their neighbours [143, 144]. A liquid instantaneous configuration, in contrast, has also negative eigenvalues of $A$, resulting in purely imaginary eigenfrequencies reflecting unstable modes.

The second step is to calculate the spectral density, $\rho(\omega)$, which is normalized to unity, $\int d\omega \rho(\omega) = 1$. In turn, we average over the normal modes of many configurations

$$\rho(\omega) = \left\langle \sum_{i=1}^{\text{dim} \ N} \delta(\omega - \omega_i) \right\rangle.$$  \hfill (7)

This density is composed of a stable part, $\rho_s(\omega)$, with real frequencies, and an unstable part, $\rho_u(\omega)$, with imaginary frequencies. The unstable part $\rho_u(\omega)$ is associated to a negative curvature in the momentary potential landscape. As described in [137–142], especially $\rho_u(\omega)$ can be related to the diffusion constant.

$$D = \frac{k_B T}{m} \int \rho(\omega) \frac{\tau_h}{1 + \tau_h^2 \omega^2} d\omega, \hfill (8)$$

and depends on the temperature $k_B T$, the particle mass $m$ and the average ‘hopping time’ $\tau_h$ for the transition across potential barriers between two local potential wells. This time is known as the inverse hopping frequency and calculated as

$$\tau_h^{-1} = c \int \frac{\omega^2}{2\pi} \rho(\omega) A \exp \left(-B \frac{\omega^2}{k_B T} \right) d\omega, \hfill (9)$$

where $c \approx 3$ is a constant taking into account the different routes to escape from a local potential minimum and the constants $A$ and $B$ are obtained from an exponential fit of $\rho_u(|\omega|)/\rho_s(\omega)$. For details, we refer the reader to [137–142].

3. Controlled change of the coupling strength by laser manipulation

3.1. General concept

In order to obtain valid information on thermodynamic properties it is essential to gain reliable control on the coupling parameter of the system. According to equation (1), three possibilities exist to control $\Gamma$: (i) controlling the charge $Q_d$ of the particles, (ii) controlling the inter-particle distance $h_{WS}$, and (iii) controlling temperature $T$. With $\Gamma \sim Q_d^2$, already a moderate variation of charge allows to change the coupling strength considerably. Thus, controlling the charge on the particles is the most tempting approach. However, this is not feasible in practice. $Q_d$ is determined by geometric properties of the particles (such as the particle radius) and by the plasma conditions in the vicinity of the particles. Except for rare special conditions, the geometric properties cannot be altered during experiments and the local plasma conditions are not solely set by the discharge parameters but are modified by neighbouring particles as well. Thus, the charge is not directly controllable by means of external parameters such as discharge power, neutral gas pressure or bias voltages. Each will affect the plasma as well as the particle arrangement and thus result in a rather complex parameter dependence. Especially the inter-particle distance is strongly determined by the mutual particle repulsion, i.e. the particle charge, and the external confinement.
Thus, there is no easy access to control charge and inter-particle distance independently. Therefore, to decrease the coupling strength, temperature is the only remaining control parameter.

3.2. Kinetic and surface temperature of dust particles

Due to the macroscopic size of the particles (at least) two temperatures have to be distinguished: the kinetic temperature $T^i$ of the particles and the particle surface temperature $T_i$, where ‘$i$’ labels different dust particles (we use the notion ‘temperature’ for the mean kinetic energy of the particle). To understand the physical difference of these temperatures we have to take a microscopic view point: The dust grains are solid bodies consisting of a very large number $N$ of molecules (their atomic substructure and electronic properties are not relevant in this context). A transfer of energy to the particle from external sources will, in general, excite all $3N$ degrees of freedom of the molecules. Among them are 6 degrees of freedom (‘centre-of-mass’ (COM) modes) which are related to a collective displacement or a rotation of all particles which leave all inter-molecular distances unchanged. In contrast, the remaining $N-6$ degrees of freedom are related to intramolecular vibrations (phonons, ‘relative modes’). It is easy to verify that the Hamiltonian of this system of $N$ molecules can be split into a centre of mass and a relative part which are independent (they commute). Therefore, only the vibrations contribute to the surface temperature $T_i$. On the other hand, the kinetic temperature (energy) is connected only to the COM modes—in fact, only to the translational degrees of freedom. Now, in case of laser heating, predominantly the COM modes are excited via the radiation pressure, see the discussion in section 1.1. This is due to the large spot size compared to the intermolecular distances (in order to excite the relative modes there should exist a substantial field gradient on the scale of the intermolecular separation which is not the case). Still, an open question is to what degree individual photons are absorbed by the molecules which could lead to an excitation of the relative degrees of freedom and, eventually, to a slight increase of the surface temperature, but this effect will be neglected here.

The surface temperature is almost the same for all particles (provided they are under the same plasma conditions), $T_i \approx T_s$, because it arises from a very large number of coupled modes of many particles $N$. In contrast, the kinetic energies of individual particles (i.e. the individual $T_i$) are, in general, different. This is because the displacements of individual dust particles caused by lasers are, to a large extent, random, as is the case with the random displacements of the molecules of a gas. And as in the case of a gas, collisions between all dust grains eventually drive the system to thermodynamic equilibrium (with respect to the collective degrees of freedom). For a classical system one would expect that a Maxwellian velocity distribution is established, the width of which then reflects the overall kinetic temperature $T$. This expectation is fully confirmed by our simulations and the laser heating experiments discussed below, see figure 5.

The decoupling of centre of mass and relative degrees of freedom and, correspondingly, of $T^i$ and $T_i$ (and, therefore also $T$ and $T_s$), has been verified—indirectly—experimentally. Here we mention detailed studies of the surface temperature by Maurer and Kersten [145] for dust particles doped with a temperature sensitive fluorescent dye. They concluded, from an energy balance model, that $T_i$ scales roughly linearly with the rf power. At low rf discharge powers, $T_i$ has been found slightly above room temperature. In contrast, in the experiments reported below, the kinetic temperature is found significantly higher than room temperature and thus it may strongly exceed $T_i$. This is because the radiation pressure from the laser(s) implies a substantial momentum transfer to the particles. Combined with their small weight and low friction in the plasma, these laser beams are capable to significantly accelerate individual dust particles.

Returning to our original goal—a control of $\Gamma$—we conclude that only the kinetic temperature has the meaning of thermodynamic temperature. (We mention that this holds rigorously only in the thermodynamic limit, however, many simulations and experiments indicate very similar behaviour in the case of finite clusters.) So, in the following we will concentrate entirely on the kinetic temperature $T$. We expect that variation of $T$ (and hence $\Gamma$) gives access to the thermodynamic properties of finite dust clusters and to phase transitions. The only requirement which we have to fulfill is that the employed heating scheme should guarantee a truly random character of the individual kinetic temperatures $T^i$. In other words, the excitation should act like a thermostat for dust particles allowing for equilibration by particle scattering. As we will see below, properly chosen laser heating fulfills this requirement very well and allows for spatially homogeneous and stationary heating.

Finally we mention that, unlike $T_s$, the kinetic temperature $T$ cannot be obtained experimentally from a conventional heat flux analysis. Recent investigations by Fisher et al [146] state that electrostatic fields in the plasma background provide a significant contribution to the kinetic temperature of dust particles. However, a complete understanding of the involved processes is still missing and this makes it difficult to control temperature this way. These questions are beyond the present review and are not crucial for our subsequent analysis. In the experiments discussed below, the kinetic temperature (kinetic energy) of individual particles is recorded directly by tracking all particles. This gives access to the collective properties of the COM degrees of freedom of the entire dust cluster, including the power spectrum and the velocity distribution.

3.3. Realizing laser heating of dust particles

The idea to transfer momentum from a laser beam to a dust particle goes back to the early days of dusty plasma research [147–152] and has been used for many purposes so far. This includes the investigation of particle interaction potentials [63, 153, 154], the excitation of waves [147, 151], study of Mach cones [90, 91, 155, 156] or the stability and normal mode analysis [53]. The first systematic laser heating experiments were performed by Wolter and Melzer [92] and by Nosenko et al [93, 94]. In the experiment of Wolter and Melzer [92] the spot of a laser beam is rapidly moved via scanning mirrors to
one position in a 2D dust cluster and remains at this position for about one tenth of a second, accelerating dust grains during this time. Then, the laser spot is rapidly moved to the next randomly chosen position via galvanometer mirrors. A similar heating scheme has been developed in Greifswald in order to manipulate finite 3D clusters [97]. There, a laser beam is used to manipulate the cluster from the horizontal direction using this ‘point and shoot’ technique. In this experiment, a near Maxwellian velocity distribution of the particles is realized, but with different kinetic temperatures in the beam direction and perpendicular to it.

The experiment of Nosenko et al [93, 94] exploits two opposing laser beams which were directed onto a 2D plasma crystal using scanning mirrors. Driving these scanners with sinusoidal signals at an irrational frequency ratio a well defined area of the plasma crystal is scanned (i.e. heated) in a Lissajous figure-like fashion. In addition, the opposing beam setup assures that the transferred momenta cancel on average, while the kinetic energy of each particle is raised as a result of non-compensated momentum fluctuations. Nosenko et al showed that their laser heating results in a Maxwellian velocity distribution parallel and perpendicular to the optical axis. However, the temperature in perpendicular direction was found to be significantly lower. Obviously, the viscous damping of the neutral gas impedes that collisions redistribute sufficient energy in perpendicular direction.

To overcome this limitation recent laser heating experiments for 2D clusters use four laser beams, where each optical axis is equipped with two opposing beams and perpendicular orientation of both optical axes [96, 157], see figure 4(a). In addition, the scanning procedure has been optimized. The reason for this is that a scanning scheme based on Lissajous figures results in a velocity power spectrum where strong harmonics of the scanning frequencies are observed. This is an indication that the periodicity of the driver causes individual particles to move with the same periodicity, i.e. between two kicks of the laser the particle velocity decreases significantly. Therefore, the optimized scanning procedure (see figure 4(b)) assures that each particle is driven by a laser beam before its motion originating from the previous laser drive is damped out. Thus, the requirement that each spatial position is covered by the scanning procedure is only sufficient if the maximum time between two complete scans is less than the inverse of the damping rate.

The experiments of Schabinski et al demonstrated that this is feasible for small clusters ($N < 100$). Their measured velocity spectra show no residual peak of the scanning lasers. Figure 5 summarizes the basic features of their heating method. In figure 5(a) the velocity distributions of the heated system are clearly Maxwellian. Plot (b) stresses that the Maxwellian character is even obtained if the velocity distribution is checked for each particle individually. The small scatter in particle temperature is a clear indication that the heating process is spatially homogeneous. Therefore, the laser heating with four laser beams and an optimized scanning procedure can be regarded as an ideal thermostat for 2D dusty plasma crystals.

4. Theoretical description of 2D laser heating

In this section, we first describe how the 2D laser manipulation experiment described above is simulated with the Langevin molecular dynamics (LMD) method [96, 157, 158] and then develop an analytical model for the achieved temperature. We explicitly study the elaborate heating scheme used in the experiments that will be presented below in section 5.2. The purpose of such computer simulations is to suggest optimal values for the heating parameters and to recommend future experiments like inhomogeneous heating. Moreover, computer simulations provide the possibility to systematically scan single parameters like the heating power or the beam size at constant other parameters which is often experimentally too costly.

4.1. Langevin Molecular Dynamics simulations

In an LMD simulation, the Langevin equations of motion for the dust component

$$m \frac{d \vec{v}_i}{dt} = \vec{F}_i + \vec{F}_{\text{int}} - \gamma m \vec{v}_i + \vec{\xi}_i(t), \quad i = 1 \ldots N, \quad (10)$$

for all dust grains $i$ are integrated numerically. In these equations, the first term on the right hand side results from the gradient with respect to $\vec{F}_i$ of the Hamiltonian, equation (3) (i.e. the forces due to external potentials and
inter-particle interactions that appear in Newton’s equations of motion). The last two terms on the right hand side describe the frequent collision of the dust grains with the neutral gas background statistically by a viscous damping force, \( -\gamma m \bar{\nu}_i \), and a random force \( \xi_i(t) \). The random force has zero average and can be modelled by a Gaussian probability distribution function with correlation function

\[
\langle \xi_{i,a}(t) \xi_{j,b}(t') \rangle = 2\gamma m k_B T \delta_{i,j} \delta_{a,b} \delta(t - t'),
\]

where \( i, j \) are particle indices and \( a, b \in \{1, 2\} \) indicate the spatial components of the random force vector. The amplitude of this force depends on the temperature \( k_B T \) as well as on the friction coefficient \( \gamma \) via the fluctuation-dissipation theorem (11).

The dust–dust interaction is described by a Yukawa potential, equation (2). In experiments on 2D clusters, the strong vertical confinement in the plasma sheath allows the formation of monolayer clusters. Therefore, the system is treated strictly 2D in the simulation.

The effect of the heating lasers on each particle is given by the second term on the rhs in equation (10), \( F_{i}^{\text{laser}} = \sum_{l=1}^{N_L} f_{i}^{l,\text{laser}} \), which is composed of separate contributions of all \( N_L \) laser beams. The dominating effect of every single heating laser beam (index \( l \)) on particle ‘i’ is a momentum transfer by the radiation pressure which is described by the force \( f_{i,1}^{l,\text{laser}} \). If the laser spot hits a dust grain, this particle is accelerated in beam direction. The laser force

\[
F_{i} = q_{\text{optic}} n_1 \frac{\pi r_p^2 I_{\text{laser}}}{c} = q_{\text{optic}} n_1 \frac{\pi r_p^2}{c} \frac{P_{\text{laser}}}{2\pi \sigma_x \sigma_y} \times \exp \left[ -\frac{\Delta_x^2}{2\sigma_x^2} - \frac{\Delta_y^2}{2\sigma_y^2} \right]
\]

is proportional to laser intensity \( I_{\text{laser}} \) at the particle position \((\Delta_x, \Delta_y)\) relative to the laser spot centre, the cross-section of the dust particle \( \pi r_p^2 \), the refractive index of the plasma \( n_1 \) surrounding the particle and a dimensionless quality factor \( q_{\text{optic}} \) [159]. This factor describing the momentum transfer by an incoming photon has the limiting values 1, if the particle was a perfect absorber, and 2 for a perfectly reflecting flat disk perpendicular to the beam. For a spherical particle reflecting, absorbing and transmitting photons it is \( q_{\text{optic}} < 2 \). Typical values of \( q_{\text{optic}} \approx 1 \) are reported for melanine particles [159]. \( \sigma_{x,y} \) characterizes the extensions of the elliptical spot which has an area of \( 2\pi \sigma_x \sigma_y \).

As sketched in figure 4, the laser beams strike the cluster from above the levitation plane with a low angle of incidence. The out-of-plane component of the accelerating force is considered to have no impact on the particles’ motion due to the strong vertical confinement. However, the spot profile is stretched in beam direction due to the relatively small angle of incidence, \( \alpha < 90^\circ \). The laser force is time and space dependent according to the experimentally chosen scheme. The amplitude of the force depends on the particle’s position inside the spot which is described by an anisotropic Gaussian intensity profile. The force acting on a particle at position \((\Delta_x, \Delta_y)\) is \( F(t) = -\Delta_x(t) - \Delta_y(t) \) away form the moving spot centre is described by

\[
\tilde{F}_i(t) = \frac{P_0}{2\pi \sigma_x \sigma_y} \cdot \exp \left[ -\frac{\Delta_x^2}{2\sigma_x^2} - \frac{\Delta_y^2}{2\sigma_y^2} \right] \tilde{e}_i,
\]

where \( \tilde{e}_i \) is a unit vector in beam direction. Here, based on equation (12), we have introduced dimensionless units for all lengths and \( P_0 \). The amplitude of the force \( P_0 = q_{\text{optic}} n_1 \pi r_p^2 P_{\text{laser}}/c \) is determined by the laser power, the cross section of the dust grain and its absorption and reflection characteristics. Since only the in-plane component has an effect on the particle’s motion, \( P_0 \) is reduced by the factor \( \cos \alpha \) where \( \alpha \) is the angle of incidence. The trajectories of the laser spot centres within the levitation plane are denoted by \( \tilde{r}(t) = \{x(t), y(t)\} \) and depend on the heating scheme.

### 4.2. Comparison of different heating schemes

All investigated heating methods (a more extensive analysis was reported in [157, 160]) use triangular signals to drive the \( x \)- and \( y \)-oscillations of the lasers spots \( x(t) = x_0 \cdot \text{triag}(f_s t) \), \( y(t) = y_0 \cdot \text{triag}(f_s t) \). Using a sinusoidal signal would cause
Especially rational scanning frequency ratios and the importance of randomly changing the scanner frequency.

For the simulation results presented in this review, we used the following dimensionless units of length, time and energy: \( r_0 = [Q_0^2/(4\pi \varepsilon_0 m_0 \omega_0^2)]^{1/3}, \ t_0 = \omega_0^{-1}, \) and \( E_0 = [Q_0^2 m_0 \omega_0^2/(4\pi \varepsilon_0)]^{1/3}. \) For the numerical integration of the stochastic differential equations, we used an integration scheme described by Mannella et al that can be considered as an extension of the ‘leap frog’ scheme [161]. For details concerning the simulations, we refer the reader to [157, 158].

As a first result, our LMD simulations confirmed the importance of randomly changing the scanner frequency. Especially rational scanning frequency ratios \( f_x/f_y \) result in a closed scanning pattern, and successful heating is achieved. A second result is the confirmation of the excellent heating qualities (with respect to homogeneity of the heating) of the random frequency method B [157]. While constant scanning frequencies and combinations of them appear as peaks in the PSD, the pseudo-irrational frequency method A-I shifts the entire heating effect to \( \sqrt{\pi} \pm \sqrt{2\pi} f = \sqrt{\pi} f \cdot \pm 1. However, in order to achieve a thermal heating effect, each particle should frequently be affected by a laser spot. This condition is violated for \( \sqrt{\pi} \) below the trap frequency \( \omega_0 \). Therefore, scanning frequencies of the order of a few \( \omega_0 \) are advantageous [157].

4.3. Analytical estimate for the temperature increase

We now derive an analytical approximation for the heating power as a function of the laser parameters: the force amplitude \( P_0 \), spot size \( \sigma_x, \sigma_y \), size of the scanned array \( 2Y \times 2Y \) and average spot velocity. The spot velocity is connected to the scanning frequencies, \( f_{x>y}, f_{y>x} \), via \( v_{x>y} = 4Xf_{x>y} \) and \( v_{y>x} = 4Yf_{x>y} \). A similar analytical derivation of the heating effect was performed by Wöller and Melzer for a setup with one laser spot that was rapidly moved to one position, remained there for a dwell time of a few microseconds and was then moved to the next position [92].

Here we extend this model to a continuously moving laser spot corresponding as closely as possible to the experimental heating method by Schablinski et al that is discussed in section 5. For simplicity, the momentum transfer is computed in one direction only. Since the velocity of the dust is small compared to the velocity of the moving laser spot, the particle’s displacement during an acceleration event can be neglected. The time-dependent force acting on a particle when a spot passes with distance \( \Delta Y_0 \) in y-direction is

\[
F(t') = \mp P_0 \frac{1}{\sqrt{2\pi}} e^{-\pi t'^2} \left[ 1 - \frac{1}{\sqrt{2\pi}} \sum_{\sigma} \exp \left\{ -\frac{1}{2} \left( \frac{\Delta Y_0^2}{\sigma_x^2} + \frac{\Delta Y_0^2}{\sigma_y^2} \right) \right\} \right],
\]

where we introduced the time scale of the passing event \( \tau = \sqrt{\frac{\sigma_x^2}{\pi^2}} + \sqrt{\frac{\sigma_y^2}{\pi^2}} \) and the shifted time \( t' = t + \tau^2 \Delta Y_0 v_{x>y} - t_0 \). Here, \( t_0 \) is the time when the spot has the smallest distance \( \Delta Y_0 \) in y-direction and we introduced the time-independent geometry factor \( \Sigma \). The total momentum transfer to the particle during this laser hit follows from equation (14)

\[
\Delta p = \int_{-\infty}^{\infty} dt' F(t') = \mp P_0 \Sigma \cdot \tau,
\]

and is cancelled, on average, by the momentum transfer from the opposing laser beam. To calculate the average energy transfer, we need to average the squared geometry factor \( \Sigma^2 \) over all possible passage distances \( \Delta Y_0 \). Assuming that the particle at \((x_p, y_p)\) does not come too close to the boundary, the integration limits can be extended to infinity, with the result

\[
\langle \Sigma^2 \rangle_{Y} \approx \frac{1}{2Y} \cdot \frac{1}{2\sqrt{\pi}} \cdot \frac{1}{\sigma_x} \cdot \frac{1}{\sigma_y} \cdot \frac{1}{v_{y>x}} \cdot \frac{1}{\tau},
\]

Each time the laser crosses the levitation plane in \( x \)-direction, it passes the particle position once. Then, the ‘passing rate’ \( \chi \) is given by the inverse crossing time in \( x \)-direction as \( \chi = \frac{\tau}{2Y} \).

The energy change of a particle with velocity \( v \) during a single laser–particle interaction event is (neglecting the deceleration due to friction during the kick),

\[
\Delta E = E' - E = v \Delta p = \frac{\Delta p^2}{2m},
\]

Table 3. Parameters defining the pattern which is scanned by the lasers. Since both beams are oriented in the ±x-direction for method A-I, \( f_x = f_y \) in that case.

| method A-I | B     |
|------------|-------|
| laser beams| 2     |
| frequency ratio | pseudo-irrational |
| \( f_x \)  | 14.5623 Hz |
| \( f_y \)  | 9.0 Hz   |
| \( f_z \)  | 50–300 Hz | 15–60 Hz |
where the momentum transfer $\Delta p$ is given by equation (15). From this the average energy transfer is obtained using equation (16),

$$\langle \Delta E \rangle = \frac{1}{2m} p_0^2 \Sigma^2 \cdot \tau^2,$$

which depends on the passing distance $\Delta y_0$ via $\Sigma$. Then, the average energy transfer per time follows from averaging over $\Delta y_0$, multiplying by $\chi$ and using equation (16),

$$\left\langle \frac{\Delta E}{\Delta t} \right\rangle = \frac{1}{2m} \cdot \frac{1}{2\sqrt{\pi}} \cdot P_0^2 \cdot \frac{1}{4XY} \cdot \frac{1}{\sigma_x \sigma_y} \cdot \tau_{eff},$$

where we also took into account that the laser spot velocity components $v_x, v_y$ vary in the experiment by computing an effective time scale $\tau_{eff}$ by averaging $\tau(v_{l,x}, v_{l,y}) = (v_{l,x}^2/\sigma_x^2 + v_{l,y}^2/\sigma_y^2)^{-1/2}$ over all possible spot velocities $v_{l,x} \in [v_{l,x}^{min}, v_{l,x}^{max}]$ and $v_{l,y} \in [v_{l,y}^{min}, v_{l,y}^{max}]$.\footnote{The integral in the expression for the effective time scale can be solved using Mathematica.}

From the result (19), we can now calculate the equilibrium temperature $T_{eq}$ in the Langevin MD model which follows from the balance between laser input power $P_{laser}$, the power input by the stochastic force, $P_{stochastic}$, and the power loss due to friction, $P_{friction}$,

$$P_{laser} + P_{stochastic} + P_{friction} = 0.$$ \hfill (20)

Assuming a 1D Maxwellian particle velocity distribution with temperature $T_{eq}$,

$$p(v) = \frac{m}{2\pi k_B T_{eq}} e^{-\frac{mv^2}{2k_B T_{eq}}},$$ \hfill (21)

and the velocity change due to friction, $\dot{v} = -\gamma v$, where $\gamma$ is the friction coefficient, we obtain

$$P_{friction} = \left( \frac{d}{dt} \frac{m}{2} v^2 \right) = -m \gamma \frac{v^2}{2} = -\gamma k_B T_{eq}. \hfill (22)$$

Using equation (20) and the power input by the laser from equation (19) multiplied by 2 to take into account the pair of lasers in each direction, we obtain our final result for the equilibrium kinetic temperature of the dust particles

$$T_{eq} = T_0 + \frac{\sqrt{\pi}^3}{2m \gamma k_B} \cdot \frac{1}{\chi} \cdot \frac{1}{XY} \cdot \frac{1}{\sigma_x \sigma_y} \cdot \tau_{eff}, \hfill (23)$$

where we introduced the laser intensity according to $I_0 = P_0/(2\pi \sigma_x \sigma_y)$. It is interesting to note the dependence on the relevant system parameters: $T_{eq}$ grows proportional to the square of the laser intensity, in agreement with the findings of Wolter and Melzer \cite{92}, and the ratio of spot area to scan area and inverse proportional to the neutral gas friction. These qualitative trends are also seen in the experiments. The time scale $\tau_{eff}$ introduces additional slightly more complex dependencies\footnote{The time scale $\tau_{eff}$ is inversely proportional to the spot velocity, when both $v_{l,x}$ and $v_{l,y}$ intervals are scaled by the same factor. Increasing the laser scanned area has the side effect of decreasing $\tau_{eff}$ via the faster spot velocity at constant scanner frequencies. Therefore, the temperature increases scales as $\Delta T = T_{eq} - T_0 \propto X^{-3}$ with the side length of the scanned area. At constant aspect ratio $\sigma_x : \sigma_y$, the passing time $\tau_{eff}$ scales linearly with $\sigma_x$.}.

Despite the simplicity of the model, the accuracy of the result (23) is very good, as is confirmed by comparison to LMD simulations, see table 4. The temperature is overestimated slightly, and the accuracy improves with $\gamma$ from about 15% to better than 10%. For very small spot sizes and low friction (not shown) the agreement is even better than 5%. For very large spot sizes, $\sigma_x \gtrsim 0.7$ in dimensionless units, the formula becomes less accurate. Then, most particles are close to the border of the scanned area compared to the spot width, violating the assumption of equation (16). Moreover, $\tau_{eff}$ becomes large in this case and the deceleration due to friction as well as the interaction with other particles during the hitting event should not be neglected.

Our result is not only in very good agreement with the simulations, it is also useful for characterizing the heating in the experiment. In fact, the spot size used in table 4 is typical...
Figure 7. PSD of the dust velocity averaged over all particles of a Yukawa cluster with \( N = 38 \) particles with different heating methods (trap frequency \( \omega_0 = 5.5 \) s\(^{-1}\)). Top: method A-I uses one pair of laser beams in the \( \pm x \)-direction and a pseudo-irregular scanning frequency ratio \( f_x/f_y \). Bottom: heating method B uses two pairs of laser beams to accelerate the particles in both \( \pm x \)- and \( \pm y \)-directions. The scanning frequencies are randomly chosen each time the border of the scanned area is reached. The spectra for \( v_x \) and \( v_y \) coincide, here. Only this method conserves the shape of the PSD by shifting the entire spectrum to higher energies.

Table 4. Comparison of equation (23) and results from a LMD simulation. Deviations refer to the value of Gamma (lines marked \( \gamma \)). The second lines show the associated temperature \( T \). For smaller spots (not listed), the agreement is even better. Parameters: particle number \( N = 25 \), screening parameter \( \kappa = 1 \), coupling parameter \( \kappa = 1 \), coupling parameter without laser heating \( \Gamma_0 = 200 \), trap frequency \( \omega_0 = 5.5 \) Hz, dimensionless laser power \( P_0 = 10 \), scanned area \( X = Y = 3.5 \).

| \( \gamma \) | Formula \( \sigma_x = 0.50, \sigma_y = 0.10 \) | Simulation | Deviation |
|----------|---------------------------------|------------|----------|
| 0.67     | 53.23                           | 63.17      | 15.7%    | \( \Gamma \) |
| 1.00     | \( 18.79 \times 10^{-3} \)       | \( 15.83 \times 10^{-3} \) | 12.5%    | \( T \) |
| 1.33     | 14.24 \times 10^{-3}            | 12.46 \times 10^{-3} | 11.4%    | \( \Gamma \) |
| 1.66     | \( 11.95 \times 10^{-3} \)       | \( 10.58 \times 10^{-3} \) | 9.60%    | \( T \) |
| 2.00     | \( 94.65 \times 10^{-3} \)       | 104.7      | 9.22%    | \( \Gamma \) |
| 2.33     | \( 10.61 \times 10^{-3} \)       | \( 9.550 \times 10^{-3} \) | 8.15%    | \( T \) |
| 2.66     | \( 118.0 \times 10^{-3} \)       | 127.7      | 7.57%    | \( \Gamma \) |
| 3.00     | \( 8.473 \times 10^{-3} \)       | \( 7.831 \times 10^{-3} \) | 6.3%     | \( T \) |

for the experiments. Assuming that the visible spot width is \( \Delta_{vis} = 3 \sigma \), our formula suggests a coupling strength of \( \Gamma^{form} = 80 \) which is in good agreement with the measured temperature \( T = 3.5 \) eV corresponding to \( \Gamma^{exp} = 63 \) for the maximum laser power used in et al [96]. However, that result has to be understood as a rough approximation, since parameters like the spot size, the trap frequency, and laser power losses in the optical setup each have an uncertainty of several percent in the experiment.

5. Experimental results for 2D dusty plasmas

We now turn to the experiments on laser heating. First, we start with 2D systems and continue, in section 6, with experiments with finite 3D dust cluster. Before presenting our experimental results for finite 2D dust clusters we briefly summarize previous work on extended systems.

5.1. Experimental results for extended systems

In many experiments in extended 2D systems the Lissajous heating scheme A-I (see section 4.2) has been applied since it allows one to densely scan (and heat) a well defined dust area. This technique has been used successfully to drive 2D extended dust systems into the liquid state [94, 165, 166]. There, it has been demonstrated that the defect concentration in steady-state laser-heating experiments exhibits an Arrhenius-type dependence on the kinetic temperature [165]. Moreover, these experiment suggest a grain-boundary-induced melting scenario which is also observed in non-equilibrium heating experiments, e.g. by changing the gas pressure of the plasma discharge [17].

Further, experiments on heat transport and particle transport properties, such as diffusion and viscosity, of laser-heated dust layers have been performed, see e.g. [89, 94, 167–170]. From these, fundamental transport parameters like the thermal diffusivity of the dust component, as well as diffusion constants, \( \Gamma \)-dependent viscosities and anomalous diffusion properties have been identified and measured. These experiments have yielded important information on the application of laser–dust interaction methods in the field of dusty plasmas which are of high relevance also for strongly correlated small dust clusters at finite temperature.

5.2. Experimental results for 2D clusters

The remainder of this section will concentrate on finite 2D clusters and their thermodynamic properties. Especially the phase transition of these small systems is of interest as it should significantly depend on the cluster size. The following experiments use the isotropic heating method B of Schablinski et al [96, 157], which has been introduced in section 4.2. As shown there, the laser heating effectively provides a heat bath for the dust particles assuring that the dust subsystem is in thermodynamic equilibrium. Changing the laser power will result in a different temperature of this thermostat. In the experiment, for each temperature, long time series are recorded to obtain the trajectories of all particles. Examples of such particle trajectories are shown for low, medium and high temperature in figure 8(a). For low temperature the particles are well localized and only a slight angular rotation is observed. At medium temperature the particles are less localized in angular direction. Finally, at high temperatures
Copyright 2013 The American Physical Society. Part (N) temperature is significantly higher than that of the less symmetric N diffusion constant for two different dust clusters. The correlation function (see section 2.4) for the experimental cluster of 19 particles. Figure 9(a) shows a highly ordered structure of the weakly manipulated cluster. The inner shell is occupied by six particles. Intra-shell neighbours are found under angles of $\varphi = 60^\circ$, 120$^\circ$ and 180$^\circ$, as is clearly seen by the peaks at these angles and a radius $r_N \approx 1$mm, corresponding to the inner shell. The outer shell is occupied by 12 particles and the angular order with respect to the inner shell is fixed. Distinct peaks are visible at multiples of $\varphi = 30^\circ$. When the laser power is increased to a moderate value (see figure 9(b)), clear intra-shell and inter-shell correlations persist. However, inter-shell rotation (‘angular melting’) has started, as the peaks at the outer radius are no longer fully separated. At the inner shell radius, weak peaks at $\varphi \approx 103^\circ$ and $154^\circ$ (arrows in figure 9(b)) indicate the occurrence of configurations with seven particles on the inner shell. The appearance of this metastable configuration indicates the onset of radial melting. Finally, at high laser power (figure 9(c)), the correlations between inner and outer shell have vanished almost completely. At this heating power, the frequent particle transitions between the two shells give rise to a finite density in the region between these shells.

Thus the C2P correlation function fully confirmed the stability of this ‘magic number’ cluster against inter-shell rotation. Yet a complete quantitative analysis of the different melting temperatures and their dependence on the particle number is still open.

6. Experimental results for 3D clusters

3D dust clusters are formed in parallel plate radio-frequency (rf, 13.56 MHz) discharges, see figure 10 and e.g. [55, 120, 173, 174]. The discharges are typically operated in argon at gas pressures between 1 and 100 Pa and at rf powers between 1 and 10 W. The dust grains trapped in the discharge generally

\[ T_M \approx 9.000 \text{ K}, \quad T_S \approx 2.000 \text{ K}. \]

A systematic investigation of melting temperatures as a function of particle number [142] has shown that symmetry has a mayor influence on melting temperatures of finite systems, confirming earlier theoretical predictions [51, 125].

The above results show that the INM analysis is sensitive to the cluster symmetry. However, it does not resolve different melting processes such as inter-shell and intra-shell melting. (For example, it is well known [51] that the above two clusters have angular melting temperatures that differ by many orders of magnitude but that their radial melting temperatures are comparable.) For this reason, we also consider the C2P correlation function (see section 2.4) for the experimental cluster of 19 particles. Figure 9(a) shows a highly ordered structure of the weakly manipulated cluster. The inner shell is occupied by six particles. Intra-shell neighbours are found under angles of $\varphi = 60^\circ$, 120$^\circ$ and 180$^\circ$, as is clearly seen by the peaks at these angles and a radius $r_N \approx 1$mm, corresponding to the inner shell. The outer shell is occupied by 12 particles and the angular order with respect to the inner shell is fixed. Distinct peaks are visible at multiples of $\varphi = 30^\circ$. When the laser power is increased to a moderate value (see figure 9(b)), clear intra-shell and inter-shell correlations persist. However, inter-shell rotation (‘angular melting’) has started, as the peaks at the outer radius are no longer fully separated. At the inner shell radius, weak peaks at $\varphi \approx 103^\circ$ and $154^\circ$ (arrows in figure 9(b)) indicate the occurrence of configurations with seven particles on the inner shell. The appearance of this metastable configuration indicates the onset of radial melting. Finally, at high laser power (figure 9(c)), the correlations between inner and outer shell have vanished almost completely. At this heating power, the frequent particle transitions between the two shells give rise to a finite density in the region between these shells.

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Figure 9. (a)-(c) C2P correlation function for a laser heated cluster of $N = 19$ particles. The low laser power corresponds to $T = 2.800$ K, the moderate (high) power to $T = 17.000$ K ($T = 34.000$ K). The first radial coordinate is averaged over the inner shell (arrow). (d) Radial pair distribution function for all three laser powers. Note that it does not distinguish between different shells. The inset shows the cluster configuration at the lowest laser power.

Figure 10. Sketch of the experimental setup to investigate laser heated 3D dust clusters. The particles are trapped in the cubic glass box and are illuminated by two Nd:YAG lasers from two sides and heated by two diode lasers from opposite directions. The three orthogonal high-speed cameras allow to trace the full 3D particle motion individually. From [175].

are monodisperse plastic microspheres with diameters chosen between 3 and 10 $\mu$m. These 3D clusters are trapped inside a cubic glass cuvette placed onto the lower electrode. The glass box provides inward electric forces on the negative dust grains. To compensate the gravitational force an upward thermophoretic force is applied by heating the lower electrode. The combination of all forces provides a 3D harmonic confinement [106]. There, the particles arrange in nested spherical shells forming Yukawa balls or Yukawa clusters [55, 120, 176], see section 2.2.

In the experiments described here, the particles are illuminated by low-intensity laser beams and the scattered light is recorded with high-speed video cameras at frame rates of 50 to 200 frames per second (fps), typically [97, 120, 176, 177]. For the observation of the 3D clusters a stereoscopic camera setup [120] is used where the particles are observed from three orthogonal directions. This setup allows to measure and reconstruct the full 3D trajectories of clusters with up to $N = 100$ particles with high temporal resolution [120, 178]. Consequently, the dynamical properties of the dust cluster can be followed for all particles individually. To realize a heating process for these 3D clusters a simpler approach than for the 2D case had to be chosen due to experimental constrains. The transfer of the elaborate heating schemes used for the 2D clusters (section 5.1) to the 3D case would require six beams, two of which would be blocked by the electrodes. Here, only two additional opposing laser beams are oriented parallel to the electrode and are randomly swept over the cross section of the cluster with a dwell time of $\tau = 0.1$ s at each position [97, 175, 177]. The two laser beams at 660 nm wavelength are operated with up to 1 W output power. The random ‘kicks’ to the particles by the radiation pressure mimic a heating process for these 3D clusters.

As a consequence, the resulting velocity distribution of the particles in the cluster is only near-Maxwellian, with an overpopulation of ‘cold’ dust particles [97]. Also the heating is more effective in the direction of the beams resulting in higher temperatures along this axis. Consequently, this laser-heating scenario of 3D clusters does not provide a true thermodynamic heating, yet. Nevertheless, from the velocity distributions reasonable kinetic temperatures $kT_\alpha = m \langle v_\alpha^2 \rangle$ with $\alpha = x, y, z$ can be assigned and values of the order of a few times $10^4$ K (few eV) have been realized by this laser heating setup.
Figure 11. Trajectories of a $N = 60$ particle cluster recorded over a time span of about 10 s. (a) Without laser excitation, (b) with 250 mW laser power, (c) Equilibrium particle positions in cylindrical coordinates $\rho = \sqrt{x^2 + y^2}$ and $z$ for the unheated case (a). From [175].

Figure 12. C2P correlation functions for the $N = 60$ cluster, (a) without laser heating ($\Gamma = 850$), (b) for a laser heating power of 100 mW ($\Gamma = 550$) and (c) 300 mW ($\Gamma = 250$). (d) Pair distribution function. The smoothening of the curve with decreasing $\Gamma$ indicates the loss of order but, in contrast to the C2P correlation function in (a)–(c), the pair distribution function cannot distinguish intra-shell from inter-shell correlations. (The zero line is shifted for $\Gamma = 550$ and $\Gamma = 250$, for sake of clarity).

Here, as an example, the heating and melting of a 3D cluster with $N = 60$ particles is demonstrated using the pair of opposing heating laser beams. The Yukawa ball is spherical in shape and consists of two shells, see figure 11. By increasing the laser power, the amount of heating is increased and melting is achieved. For low laser heating power the cluster remains in a solid-like arrangement, as seen from the particle trajectories. Stronger random particle motion is excited at higher laser power where then frequent intra-shell and inter-shell particle exchanges are seen. Hence, the cluster is apparently driven into the liquid regime. The change of structural and thermodynamic properties that is induced by the lasers is illustrated in figure 12(d). There we show the pair distribution function $g(r_{ij})$ for three different heating powers. The differences in $g(r_{ij})$ between the three heating powers are only small. All three curves show a pronounced first-neighbour maximum at $r_{ij} \approx 0.6$ mm, a shoulder at $r_{ij} \approx 1.2$ mm corresponding to second neighbours and a decay to zero. While the curves for $\Gamma = 250$ and $\Gamma = 550$ appear smooth, a substructure is visible at high coupling, $\Gamma = 850$. The reason for the weak sensitivity is that $g(r_{ij})$ does not distinguish between intra-shell neighbours and neighbours on different shells. This leads to a smeared out structure of $g(r_{ij})$.

The loss of order is reflected more clearly in the C2P correlation function. As for the 2D case, it allows to quantitatively assess the melting transition(s) for these 3D clusters.

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clusters. This is shown in figure 12 for the same cluster \((N = 60)\) with or without laser heating. As for the 2D case, one sees pronounced peaks at distinct angles \(\varphi\) and radii \(r_{\Omega}\) at low heating powers and subsequent loss of correlations for increased heating (reduced \(\Gamma\)). Also, in these 3D systems, laser heating provides a near-equilibrium heating scenario. A two-step melting (orientational melting before radial melting) which is expected for finite clusters [51, 52, 126, 179] is experimentally identified also for 3D clusters [97].

Finally, as another example of a liquid state property, the diffusion constant \(D\), equation (8), has been derived from the experimental 3D particle trajectories of the \(N = 60\) cluster, based on the analysis of unstable INMs [142, 171, 177], see section 2.5. The so obtained diffusion coefficient is shown in figure 13. For comparison, also the diffusion constants of clusters of different sizes are added [177]. A linear relationship between \(D\) and temperature is found for all studied clusters where \(D\) reaches values of about \(D = 1.3 \times 10^{-6} \text{ m}^2/\text{s}\) at the highest dust temperatures, \(T \approx 4 \times 10^4 \text{ K}\). The values for the diffusion coefficient are decisively larger than in the 2D case [142, 171]. A reason for this is the higher dimensionality of the system that allows more paths to change configurations\(^{15}\).

As discussed in section 5.2, from the diffusion constant an approximate melting temperature can be extrapolated which is then found to be \(T_M \approx 2010 \text{ K}\) for \(N = 60\). Interestingly, this is smaller than the kinetic temperature of this cluster even in the absence of heating which is \(T_{\text{kin}} = 2930 \text{ K}\) [177]. This relatively high kinetic temperature, even in the unheated case, is explained by additional heating processes by the wake-field instability caused by streaming ions in the plasma sheath [180–182].

7. Discussion and outlook

7.1. Discussion of the results

In this review, we discussed the various opportunities provided by laser beams as manipulation tools for dusty plasmas. We focused on the use of lasers as heating instruments for dust particles. As discussed in the introduction, various further uses, like the excitation of shear stress [88, 89] or of the rotation of a cluster shell [53], are possible as well, but this goes beyond the scope of this review. The availability of reliable heating tools is essential for the experimental investigation of phase transitions and instabilities in dust crystals. The enhanced manipulation setup for finite 2D dust clusters, presented in section 3.1, has proven to be usable as a tunable thermostat in both experiment and LMD simulation. The experiments confirmed that the used laser heating scheme provides a homogeneous power input over the entire cluster and over all frequencies. As desirable for a true thermal heating, the isotropy and the Maxwellian shape of the velocity distribution are preserved to very high accuracy. Various laser scanning concepts have been studied in the simulations allowing to predict the optimal parameters for the experiments.

\(^{15}\)From the relation of the diffusion coefficient to the mean squared displacements, equation (5), one would expect a dimensionality-related increase by a factor \(3/2\) which, however, is not sufficient to explain the observed difference of the experimental values. This problem is still under investigation.

The laser heating method was used to perform temperature scans of small 2D dust clusters with different particle numbers. The INM analysis allowed us to calculate the diffusion constant and, by this, to determine an approximate melting temperature. This melting temperature is found to be crucially dependent on the exact particle number, as a consequence of different cluster symmetries. At the same time the INM analysis does not allow to discriminate between intra-shell and inter-shell melting. As another more sensitive quantity we studied the C2P correlation function which displays both intra-shell and inter-shell correlation.

The heating method for 3D dust clusters has to work with two opposing laser beams, due to the required space for diagnostics and illumination of the cluster. Hence, the heating quality with respect to the isotropy and the Maxwellian shape of the velocity distribution is not as perfect as for 2D clusters. Nevertheless, as in 2D, the quality of this heating technique is sufficient to manipulate the dust temperature (and hence the coupling strength) in a controlled manner, without affecting the plasma parameters. Due to the spherical symmetry of 3D Yukawa balls, structural parameters which take into account this symmetry are required in order to investigate the structure and the melting behaviour of these systems. The most sensitive quantity turned out to be the C2P correlation function which displays both intra-shell and inter-shell correlation. This correlation function should allow to study these questions in detail in the near future.
Both experiments and LMD simulations have shown that an appropriate heating scheme allows for a homogeneous heating of the entire 2D cluster [96, 157]. However, this heating method can also be used to heat only a selected spatial region. As long as the heated region is larger than the laser spot size, local heating is simply realized by restricting the area which is scanned by the laser spots. Kudelis et al suggested an experiment where only the central region of a 2D Yukawa cluster is heated by four randomly moving laser spots and performed LMD simulations for this scenario [158]. The thermal conductivity associated with the radial temperature profile, see figure 14 (bottom), is found to be constant over a wide range in coupling strengths, including the phase transition between solid and liquid [158]. This result is in good agreement with experimental data for the heat transport in an extended 2D dust crystal by Nosenko et al [94]. The upper part of figure 14 shows the spatial particle density of the Yukawa cluster that is heated in the inner region. While a pronounced shell structure is found at the cold outer region, the centre of the cluster is molten.

It remains an interesting task to verify these predictions in an experiment and, thus, employ laser heating for the measurement of heat transport in strongly correlated finite dusty plasmas. Another interesting task is to study the influence of an electric or magnetic field on the transport coefficients.

The applications discussed so far all considered stationary states. However, the laser heating method also allows for the study of transient, time-dependent processes. Compared to the relatively slow time scale of the heavy dust particles, the power input can be turned on and off practically immediately by switching on an off the external laser source. A possible application is a temperature quench during which the system is abruptly cooled (or heated). This allows, for example, to study the time-dependent relaxation from a fluid into a crystalline state. Alternatively, quenching of a dust crystal was achieved by changing the properties of the ambient plasma, e.g. via a sinusoidal modulation of the dc self-bias of the lower, powered electrode [183], or by applying shock waves induced via a wire placed beneath the crystal [184, 185]. However, this alters the plasma environment and does not allow to vary only $\Gamma$ by a single parameter, namely the dust kinetic temperature, like in the laser experiments presented in this review.

Feng et al [166] laser-heated, and subsequently rapidly cooled, a 2D dust crystal that initially was arranged in a hexagonal lattice. During rapid heating, the dust arrangement remained in a solid structure at temperatures above the melting point, demonstrating solid superheating. The relaxation of small, 2D dust clusters was investigated by means of lasers by Lisin et al [186]. There and in [184], the cooling rate for the crystallization of a 2D dust system was found to be close to the friction coefficient, at moderate damping rates. A different behaviour was found in laser-mediated recrystallization experiments with small 3D dust clouds [187] where the cooling rate was found to be decisively lower than the friction coefficient for moderate damping (i.e. $\omega_0 \approx \gamma$), confirming previous simulations [188]. However, further investigations are required to study the shell formation process in larger dust clouds with several thousand particles and a multi-shell structure. A possible approach to circumvent the obstacle of high particle numbers could be the use of tracer particles to artificially reduce the particle density [189, 190].

Furthermore, since dust clusters are, at low neutral gas pressure, strongly influenced by wake effects due to fast streaming ions and thus exhibit attractive forces leading to particle chain formation [63, 64], it would be tempting to apply a laser-induced torque to the dust system in order to study the energetic landscape by screwing the aligned dust ensemble.

A particularly interesting application would be a temperature quench in the presence of a strong magnetic field. Ott et al found in MD simulations that a magnetic field may prevent crystallization. This is unexpected since, due to the Bohr von Leeuwen theorem, a magnetic field should not affect the static properties of a classical system. The reason for the observed effect is that a strong magnetic field may prevent
conversion of potential energy into kinetic energy creating a bottleneck for a phase transition. Above a critical magnetic field strength, the relaxation time \( \tau \) for the crystallization increases exponentially [191]. It would be very interesting to verify these simulation results in a laser heating experiment.

So far magnetizing dust particles in an experiment has not been possible which is due to the low specific charge \( Q_d/m \) of the dust grains. An alternative way to effectively ‘magnetize’ the dust component is to put the cluster into rotation via a rotation of the neutral gas [192]. The Coriolis force then has the same functional form, \( \vec{F}_C \propto \vec{v} \times \vec{\omega} \), as the Lorentz force and acts as a ‘pseudo-magnetic’ field (Larmor’s theorem). This idea has in fact been realized in dusty plasma experiments. It was shown that important properties such as collective modes of magnetized strongly correlated plasmas can be accurately reproduced [193–195]. In combination with the laser heating (or cooling) method presented in this paper, this technique should allow to perform temperature quenches in experiments with finite dust clusters and to investigate the influence of the magnetization on the relaxation time for crystallization.

This outlines just a few possible directions of laser heating of strongly correlated finite dust clusters. Besides thermodynamic properties which were in the focus of this article, an experimental analysis of time dependent processes is now within reach. The relevance of correlation effects in many fields of physics should make such studies interesting also beyond the field of dusty plasmas.

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