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Melting and heating of two-dimensional Coulomb clusters in dusty plasmas

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The heating and melting of two-dimensional dust clusters with one additional particle in the lower layer has been investigated experimentally in a gas discharge. The full dynamical properties of the system during the entire phase transition were determined in terms of the spectral power densities of the crystal modes. A two-step melting transition is identified when the gas pressure in the discharge is reduced: first, a sudden increase of the dust temperature takes place due to an instability of the lower-layer particle resulting in a hot crystalline state of the cluster, and second, the actual transition into a fluid state is observed at a decisively lower gas pressure.

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Dusty plasmas are ideal systems to study the dynamics of plasmas as well as condensed matter on a microscopic kinetic level since the trajectories of individual particles can be measured with high temporal and spatial resolution [1]. One of the most interesting and challenging questions, however, is associated with the dynamics of phase transitions in these systems.

In dusty plasmas, micrometer sized spherical dust particles are immersed in a gaseous plasma environment. The microspheres are charged negatively to several hundreds or thousands of elementary charges by the continuous inflow of plasma electrons and ions. Usually, the particles are trapped in the sheath of a gas discharge where the electric field force on the dust balances their weight. There, the microspheres arrange in horizontally extended, nearly two-dimensional (2D) systems with 1 to 10 layers. Since the electrostatic interaction of neighboring particles by far exceeds their thermal energy the microspheres crystallize into ordered arrangements.

Monolayer systems can undergo a phase transition when the particle density exceeds a threshold value [2]. Plasma crystals with two or more layers show a nonequilibrium melting transition when the gas density in the discharge is reduced [3,4]. The phase transition can be explained by an instability due to nonreciprocal attractive forces between the particles of the different layers [5]. The attractive force is a result of the supersonic ion flow in the plasma sheath. The ion flow results in the formation of a positive space charge, the "ion focus," beneath the particles. Since the flow is supersonic the formation of the ion focus can only be communicated to the downstream particles and there is no reaction on the upstream particles. Thus the downstream particles are attracted by the ion focus, but the upstream particles are not [6,7].

From linear theoretical analysis [5] the nonreciprocal attraction leads to an oscillatory instability when the gas friction, i.e., gas pressure, is reduced below a threshold value. Then the vertically aligned particles perform horizontal oscillations with growing amplitudes which leads to an increase of the particle kinetic energy. The oscillations and the heating have been observed in experiments [3,4]. Detailed nonlinear simulations of extended two-layer and multilayer systems [8,9] have predicted that the phase transition occurs in two steps with reduced gas friction. In the first step, the crystal is heated by the oscillatory instability, but no melting takes place. The system is in a hot crystalline state. Only in a second step at further reduced friction, the crystal undergoes the melting transition into an unordered system.

In this paper, we present experiments that reveal the full dynamic properties of the melting process of (finite) plasma crystals. The system conceived for this purpose consists of a single-layer crystal with a finite number of particles, a so-called 2D Coulomb cluster, where one additional particle intentionally was placed in the second layer (see also Fig. 1). The advantages of such a system are that, first, the heating effect can be definitely attributed to the single lower-layer particle, and, second, the full dynamics in terms of the normal modes of the cluster is accessible [10]. We have experimentally confirmed that single-layer clusters without a second-layer particle do not show a melting transition, they are stable in the range of gas pressures that are of interest here.

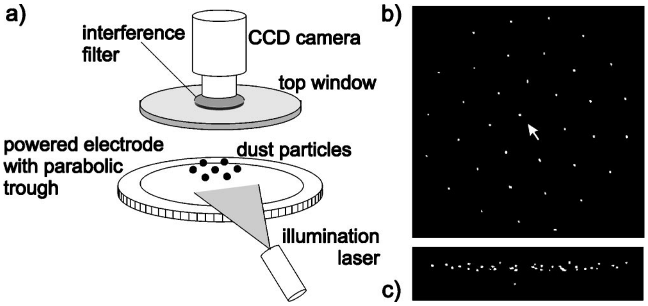

FIG. 1. (a) Scheme of the experimental setup, (b),(c) snapshots of a cluster with $N=36$ upper particles and a single lower particle. (b) Top view. (c) Side view. The side view camera is slightly titled with respect to the cluster plane, so the upper layer appears as an elliptical disk.

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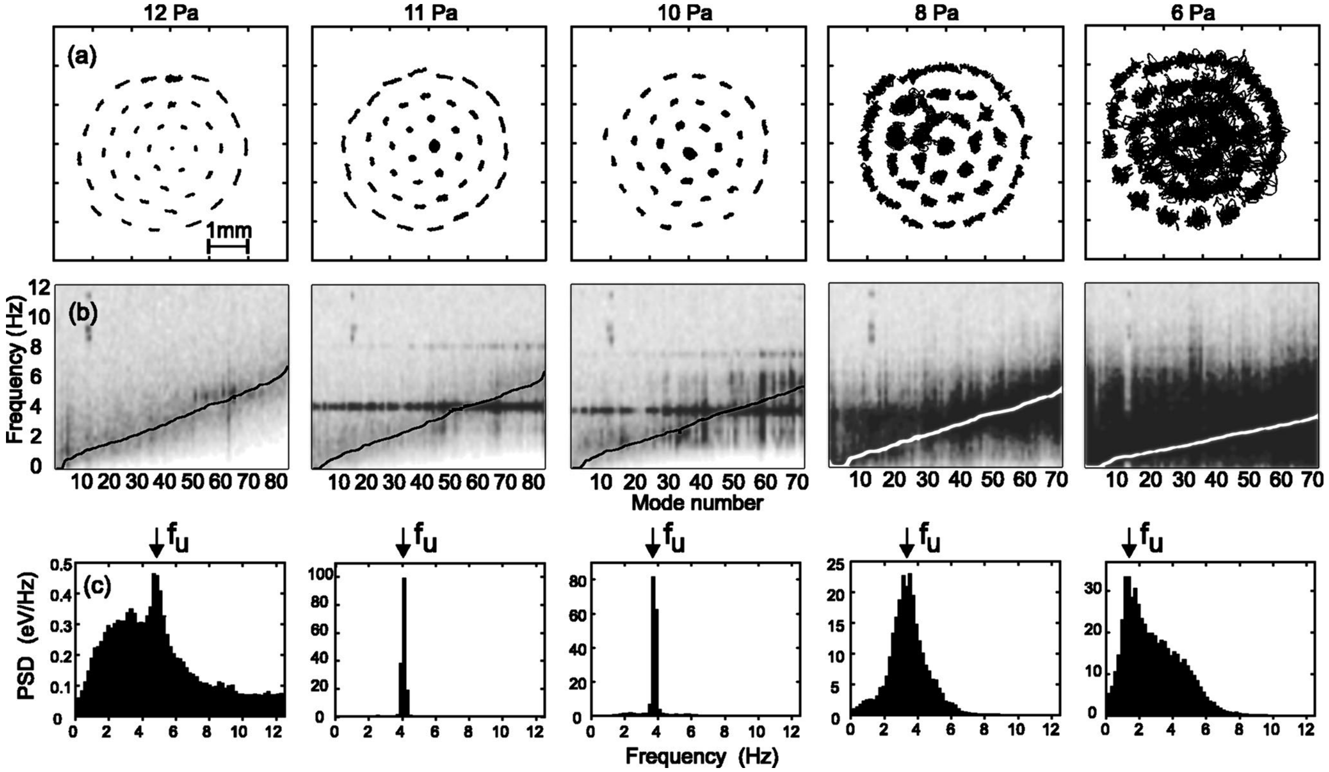

FIG. 2. Melting transition of the dust cluster with decreasing gas pressure. (a) Trajectories of the cluster particles. (b) Grayscale power spectra of the normal mode oscillations of the cluster. Darker colors correspond to higher power density. The theoretical mode frequencies for a solid state are also indicated as the black or white solid lines. (c) Power spectral density (PSD) integrated over all mode numbers. Note the drastically different scales of the vertical axes.

The experiments have been performed in a capacitively coupled rf discharge operated in argon, see Fig. 1(a). The lower electrode was powered at 13.56 MHz and 2 W. Melamine-formaldehyde microspheres of 9.55 μm in diameter (and a mass of $m_d = 6.90 \times 10^{-13}$ kg) are dropped into the plasma. The horizontal confinement for the particles is realized using a shallow circular parabolic trough in the electrode. Microspheres were dropped into the discharge until the desired cluster configuration was obtained. The microspheres of the upper layer are illuminated by a laser sheet and the particle motion is recorded with a video camera from the top for typically 41 s, corresponding to 2048 frames at 50 frames per second. In the experiment the gas pressure was varied between 4 and 15 Pa to cover the entire range of the melting transition from fluid to solid.

The dynamical properties of the dust system have been extracted from the thermal Brownian motion of the microspheres, a technique that has been applied to extended 2D systems [11] and 2D dust clusters [10]: first, the $2N$ eigenmodes of a cluster with N particles in the upper layer are calculated from the experimental configuration [12]. Then, the contribution of the particles' thermal velocity to each mode number $\ell = 1, \ldots, 2N$ is identified. Finally, from that the power spectral density (PSD) of each mode $S_\ell(\omega)$ is calculated. This way, the PSD of all modes of the cluster are determined from a single video sequence [10].

Figures 1(b) and 1(c) show the cluster with $N=36$ particles in the upper layer and a single particle in the lower layer. The lower-layer particle stays vertically aligned with a central upper-layer particle [marked with the arrow in Fig. 1(b)] at high gas pressures (12–15 Pa). With decreasing gas pressure, the lower layer particle starts to oscillate about its vertically aligned equilibrium position due to the instability arising from the nonreciprocal attraction. Due to the mutual Coulomb repulsion, the oscillating lower particle heats the upper particles. At the lowest gas pressures (below 8 Pa) the lower particle is still oscillating below upper layer particles, but from time to time this particle may jump from one upper particle to another thus heating different particles. A similar particle behavior is reported in [13]. Here we have exploited the lower particle to induce the phase transition in the upper layer.

For illustration of the melting transition of the dust cluster, the particle trajectories are shown in Fig. 2(a). At the highest gas pressure (12 Pa) the particles only slightly move around their equilibrium positions, at reduced pressure (10 to 11 Pa) the oscillations become visible from the circular particle trajectories. At lower pressure the particles start to exchange equilibrium positions which is an indication of melting (at 8 Pa an exchange has nearly occurred, at 6 Pa frequent exchanges take place).

As one major result, the dynamics of the cluster melting process is revealed in great detail by analysis of the power spectra, see Fig. 2(b). For 12 Pa, the PSD of the individual modes is concentrated around a quite narrow band of frequencies that closely follows the mode theoretical frequencies of a solid cluster with a particle charge of $Z = 11\,000 \pm 1000$ and a screening length of λ_D

=(1000±500) μm. With reduced gas pressure, the spectrum is changed completely. All modes show a maximum at the same frequency $f_u=4$ Hz which is apparent from the dark horizontal band in the spectrum. This frequency corresponds to the unstable oscillations, as will be discussed below in more detail. The dominance of this frequency in *all* modes is clearly surprising. In addition to this dominant frequency, the underlying mode structure of the crystalline state is still faintly observable in the spectrum. Below 8 Pa the situation changes again. The spectrum becomes broad for all modes and the close relation to the solid-state mode frequencies is lost. From this, it might be suspected that the cluster is in a liquid state.

The situation becomes more obvious from the mode-integrated power densities in Fig. 2(c). At high gas pressures (12 Pa) the PSD is small in absolute units and covers a broad frequency range, as expected for the density-of-states of a crystal. In addition, a small peak is observed at about $f_u=5$ Hz. This peak is a manifestation of the oscillatory instability although the instability is still suppressed at this high pressure, as explained below. At the reduced pressure of 10 and 11 Pa the PSD collapses to a single sharp peak. This definitely shows the dominance of the unstable oscillation. Then, below 8 Pa, the spectrum becomes very broad again. The spectral density is much larger (in absolute units) than in the crystalline case. The peak associated with the unstable oscillation is reduced to $f_u=3.4$ Hz (8 Pa) and 1.5 Hz (6 Pa). Similarly, the plasma frequency $2\pi f_{\mathrm{pd}}=(Z^2e^2/4\pi\epsilon_0 ma^3)^{1/2}$ reduces from $f_{\mathrm{pd}}=6.1$ Hz (12 Pa) to $f_{\mathrm{pd}}=3.4$ Hz (6 Pa) due to an increase of the mean particle distance from $a=670$ μm to $a=930$ μm.

The observed frequency f_u in the spectra is exactly that of the unstable horizontal oscillations of the vertically aligned pair, which we also have confirmed using the side view camera. The unstable oscillations of the vertically aligned pair set in at 11 Pa. The oscillating vertical pair forces all particles in the upper layer to oscillate at this frequency. This leads to the very sharp peak observed in the PSD. However, the particles do not oscillate coherently. A coherent motion, like radial, rotational, or linear in x or y direction, would result in a single or a few modes with high spectral power density which would be readily seen in the mode resolved spectrum. The fact that *all* modes are excited with similar energy indicates an incoherent motion of the particles, but at the single frequency f_u.

With reduced pressure the oscillations of the aligned pair become more violent, driving stronger oscillations in the upper particle cloud until the cluster enters the liquid state at 8 Pa. In the liquid state, the interparticle distances are not sharply defined and the corresponding spectra as well as the peak of the unstable oscillations get broader in the frequency space.

In the solid state at 12 Pa, the unstable oscillation already manifest in the spectrum although the instability sets in at a lower gas pressure. Obviously, the instability already contributes to the thermal Brownian fluctuations above the threshold pressure. Similar observations have been made in other types of nonequilibrium systems [14]: Near the instability threshold, thermal fluctuations close to the unstable frequency have a longer decay time than those far from that frequency. Thus the unstable frequency becomes preferred in the spectrum. Consequently, those modes that have a normal mode frequency close to the unstable frequency, i.e., modes numbered 50–65 in the mode resolved spectra [Fig. 2(b)], show an increased spectral power density. This illustrates the power of the applied PSD technique.

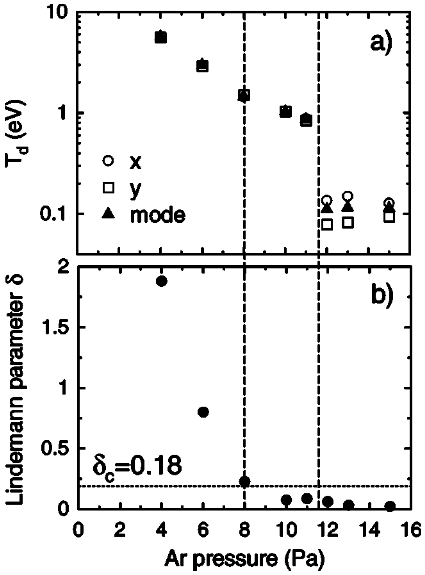

FIG. 3. (a) Dust temperature T_d (derived from the energy of the modes and from the velocity fluctuations in the x and y direction) and (b) Lindemann parameter δ as a function of Ar pressure.

The dynamical analysis has clearly revealed three distinct regimes with reduced pressure, first, a crystalline state, second, an oscillatory, but still crystalline state, and third, a fluid state.

This finding is substantiated by the measurement of the global parameters dust temperature T_d and Lindemann parameter δ. The dust kinetic temperature can be measured from the energy stored in each mode ℓ via $N\,kT_{\mathrm{mode}}=\Sigma_{\ell=1}^{2N}(1/2)m\int S_\ell(\omega)d\omega$. Alternatively, it is derived from the root-mean-square fluctuations of the particle velocity in the x and y direction, i.e., $(1/2)kT_{x,y}=(1/2)m\langle v_{x,y}^2\rangle$. The three dust temperatures are shown in Fig. 3(a). The Lindemann parameter measures the averaged relative displacement of the particles from their equilibrium positions using

$$\delta^2=\frac{1}{Na^2}\left\langle\sum_{i=1}^{N}\frac{1}{N_b}\sum_{j=1}^{N_b}|\vec{u}_i(t)-\vec{u}_j(t)|^2\right\rangle, \qquad (1)$$

where N_b is the number of the neighbors to particle i, and $\vec{u}_i$ is the relative displacement of particle i from its equilibrium position. The Lindemann parameter indicates the order in the system and is given in Fig. 3(b). Melting of 2D systems occurs when the Lindemann parameter exceeds the critical value $\delta_c=0.18$ [15,16].

At the first transition, the upper particles are dramatically heated from $T_d\approx 0.1$ eV to $T_d\approx 1$ eV. Then the temperature progressively increases as the pressure is further reduced. The temperature jump is exactly associated with the onset of the unstable oscillations. The increased temperature, however, does not lead to melting which is seen from the Lindemann parameter that stays at small values until the gas pressure is reduced to 8 Pa, where the Lindemann parameter is found to be $\delta = 0.23$ which is slightly larger than the critical value. Only then a sudden increase of the Lindemann parameter is observed which indicates melting. Thus the jump in temperature and the melting transition occur at distinctly different gas pressures.

The source of the unstable oscillations, and thus the heating and melting, lies in the vertically aligned pair of particles. We have confirmed that under the conditions of our experiments clusters without the lower-layer particle do not show the instability and do not melt. This finding excludes heating mechanisms that involve changes of the plasma properties only [17]. A *vertical* coupling of the aligned pair would involve the resonance frequency of the vertical potential well which is of the order of 15–20 Hz in our experiment. Although the camera is able to resolve these frequencies at 50 frames per second, a signature of these high frequencies was not observed.

Simulations of infinite two-layer 2D plasma crystals with nonreciprocal attraction have predicted a similar two-step melting scenario [9]. Our system is different from the simulations by having only a finite number of particles in the upper layer and only a single particle in the second layer. Nevertheless, the characteristics of the experimental PSD as well as the behavior of the dust temperature and the Lindemann parameter are very close to the findings from the simulations. This strongly supports the nonreciprocal attraction due to the ion flow as the origin of the heating and the melting of the dust cluster.

Summarizing, the detailed dynamical processes of the nonequilibrium melting of 2D Coulomb clusters in dusty plasmas have been determined experimentally. Using a single-layer system with one lower-layer particle we were able to resolve the dynamics of the cluster during the entire melting transition. The use of only a single second layer particle allows one to pinpoint the origin of the melting to the nonreciprocal attraction. The melting involves two stages of transitions in physical quantities: first, a sudden increase of the dust temperature due to the onset of the unstable oscillations, and second, a jump of the Lindemann parameter (corresponding to the actual melting transition) at a lower gas pressure. Accordingly the dynamics of the system changes from a broad solidlike spectrum, first, into a very peaked spectrum of an oscillating system, and second, into the broad spectrum of a fluid state.

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