Particle temperature measurement using pair distribution function in complex plasmas

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Abstract. In order to obtain more reliable particle temperature of complex plasmas, a new method of obtaining it in a solid phase is investigated. Since a conventional method uses a velocity distribution function (VDF), both high spacial resolution and sufficiently large observation area are required for sufficient measurement accuracy. It is often difficult, however, to satisfy them simultaneously due to necessities of highly magnified observation for the former requirement and low magnified one for the latter one. Therefore, we investigate another method of estimating particle temperature in the solid phase by use of a pair distribution function (PDF). This method has a feature that only one observation magnification is required. From comparison between the experimentally obtained PDF's and calculated ones, mean square displacement (MSD) is determined. The temperature is estimated by using the MSD if the particle motion is regarded as harmonic oscillation. It is found that the obtained temperature from the PDF is consistent with that from the VDF.

1. Introduction
Solid phase formation in complex plasmas was predicted in 1986 [1]. After that, many laboratories begun researches of observation of the solid phase. In 1994, the solid phase was successfully observed [2, 3, 4, 5]. Today this solid phase is called the Coulomb crystal or Plasma crystal.

There are many scientific issues in the complex plasmas researches, for example, Coulomb crystal formation mechanisms, the critical point in charged systems, particle and power balances, morphology and equilibrium form of the Coulomb crystals, solid-solid phase transition and so on. Since the particles usually have equally negative charges, it is often considered that the Coulomb Crystal is similar to the Winger crystal [6]. In addition to the Coulomb repulsive force, however, other forces working as attractive forces have been reported [7, 8, 9, 10]. It is considered that some attractive forces such as the ion drag force [10] are large enough to affect the crystal morphology. If particle kinetic energy, or temperature, is large enough to flee from the attractive forces, the morphology may be changed.

The critical point in charged systems has been predicted [11] though is may be difficult to observe it in usual plasmas. The complex plasmas may be suitable for the observation
and thus theoretical investigations have been carried out [12, 13, 14, 15, 16]. Since the particle temperature is one of the most important parameters for the critical point, the precise temperature measurement is necessary. Investigations on the particle and power balances also require it.

The temperature is usually estimated by using a velocity distribution function (VDF). The VDF requires sufficiently large number of particles in an observation area to get statistical reliability. On the other hand, velocity resolution depends on spacial resolution. This means that the high spacial resolution is required, especially for a small velocity region. It is often difficult to satisfy these requirements simultaneously.

Therefore, we investigate another method to obtain the particle temperature by using a pair distribution function (PDF). The PDF includes the temperature effect. This effect is called the temperature factor or the Debye-Waller factor, \( \exp (-M) \). In this paper, the experimentally obtained PDF, PDF\(_{exp}\), is compared with the calculated PDF, PDF\(_{cal}\), to determine the temperature factor. The PDF\(_{exp}\) is obtained from the two-dimensional particle coordinates based on the ground-based experiments in this paper. The PDF\(_{cal}\) is also calculated from the two-dimensional coordinates with a certain deviation of a normal distribution. From the determined temperature factor, the mean square displacement (MSD) is obtained. Thus the particle temperature is estimated from the MSD by assuming the harmonic oscillation of the particles. Although the new method is applicable to the temperature measurement in the solid phase at present since we assume the harmonic oscillation, this is still useful because the crystalline is easily formed on the ground in many experimental conditions. We also discuss the temperature estimated from the PDF by comparison with that from the VDF.

2. Theory for temperature measurement

General expression of the pair distribution function (PDF) of a Coulomb crystal in three-dimension is described as

\[
g(r) = \frac{1}{V \rho} \sum_h \sum_k \sum_\ell F_{hk\ell} \exp \left\{ 2\pi i (hx + ky + \ell z) \right\}, \tag{1}
\]

where \( V \) is volume of a unit cell, \( \rho \) average number density of particles and \( F_{hk\ell} \) the structure factor. The variables \( h, k \) and \( \ell \) are the Miller indices, \( x, y \) and \( z \) the positions in the unit cell and \( r = \sqrt{x^2 + y^2 + z^2} \). The structure factor is described as

\[
F_{hk\ell} = \sum_m \exp (-M) \exp \left\{ -2\pi i (hu_m + kv_m + \ell w_m) \right\}, \tag{2}
\]

where \( u_m, v_m \) and \( w_m \) the coordinates of the \( m \)-th particle in the unit cell. The term of \( \exp (-M) \) is called the Debye-Waller factor or the temperature factor. This is expressed as

\[
\exp (-M) = \exp \left[ -2\pi^2 \left\{ \sigma_{as}^2 + \left\langle (\Delta x)^2 \right\rangle \right\} \left( h^2 + k^2 + \ell^2 \right) \right], \tag{3}
\]

\( \sigma_{as} \) is the standard deviation of the atomic scattering factor, \( \left\langle (\Delta x)^2 \right\rangle \) the MSD. Please pay attention to the following two points in Eqs. (2) and (3). One is that the atomic scattering factor should not appear in this equation since the particles are completely classical. The other one is that the extinction rule does not work due to direct observation of the particles. This means that the summation of \( -2\pi i (hu_m + kv_m + \ell w_m) \) should be replaced to \( \rho V \). Therefore, the PDF for the complex plasmas should be expressed as

\[
g(r) = \sum_h \sum_k \sum_\ell \exp \left\{ -2\pi^2 \left\langle (\Delta x)^2 \right\rangle \left( h^2 + k^2 + \ell^2 \right) \right\} \times \exp \left\{ 2\pi i (hx + ky + \ell z) \right\}. \tag{4}
\]
Although the PDF can be obtained from Eq. (4) in principle, it is not easy to calculate it due to slow convergence of the summation on the Miller indices. The better calculation method for the complex plasmas exists, that is, direct calculation from the coordinations of test particles. This method is not applicable to an actual crystal but to the Coulomb crystal due to visual sizes of the particles. The PDF is easily calculated from the coordinates though sufficiently large number of particles is required for statistical reliability. The three-dimensional PDF is described as

\[
g(r) = \frac{4}{3} \pi r_{\text{max}}^3 \sum_{i=1}^{N_i} N_j (r - \Delta r/2, r + \Delta r/2) \frac{N_j}{N_j N_i^\text{total}} 4\pi \left\{ (r + \Delta r/2)^3 - (r - \Delta r/2)^3 \right\}
\]

\[
ge(r) = \frac{r_{\text{max}}^3 \sum_{i=1}^{N_i} N_i (r - \Delta r/2, r + \Delta r/2)}{3 N_j N_i^\text{total} r^2 \Delta r},
\]

where \(r_{\text{max}}, N_i, N_j,\) and \(N_i^\text{total}\) are the radius of the PDF calculation area, the number of particles in a spherical shell from \(r - \Delta r/2\) to \(r + \Delta r/2\), the number of calculation areas and the total number of particles in one calculation area, respectively. The two-dimensional PDF is similar formula to the three-dimensional one like

\[
g(r) = \frac{r_{\text{max}}^2 \sum_{i=1}^{N_i} N_i (r - \Delta r/2, r + \Delta r/2)}{2 N_j N_i^\text{total} r \Delta r}.
\]

In this case, the shell is not spherical but concentrically circular. In the ground-based experiments, the Coulomb crystal are usually observed as two-dimensional crystals. Actually, the Coulomb crystal observed in our apparatus typically has a few layers. Therefore, we use Eq. (6) to calculate the PDF in this paper.

To calculate Eq. (5) or (6), the particle coordinates are needed. In this paper, to set the particle at a certain position, we assume that probability of the displacement \(\Delta x\) from a lattice point is described by a normal distribution like

\[
f(\Delta x) = \frac{1}{\sqrt{2\pi} \sigma} \exp \left\{ -\frac{(\Delta x)^2}{2\sigma^2} \right\}.
\]

where \(f\) and \(\sigma\) are the probability and the standard deviation, respectively. In this case, the MSD is expressed as

\[
\langle (\Delta x)^2 \rangle = \int_{-\infty}^{\infty} x^2 f(x) \, dx = \sigma^2.
\]

Therefore, the square root of MSD is equal to the standard deviation.

The Box-Muller transform [17] is a well-known method of generating random numbers satisfying the normal distribution. Thus the particles are set at the positions determined from this method. The obtained two-dimensional particles are shown in Fig. 1 (a). The dashed triangular lines and closed circles in this figure represent perfect lattices and set particle with \(\sigma\) of 0.1, that is, \(\langle (\Delta x)^2 \rangle = 0.01\). The elementary lattice in this case is an equilateral triangle. It is confirmed that some particles are close to the lattice point, while some particles are far away from it.
Figure 1. Particles with displacement obeyed by the normal distribution and calculated pair distribution function; (a) example and (b) PDF. The dashed lines and closed circles in (a) represent ideal lattices and particles, respectively. In this case, the standard deviation of the normal distribution is 0.1.

The MSD is closely connected to the Lindemmann melting law, that is,

$$\chi = \frac{\sqrt{\langle (\Delta x)^2 \rangle}}{a},$$

where $\langle (\Delta x)^2 \rangle$ is the MSD and $a$ is the lattice constant. It is known that the value of $\chi = 0.3 \sim 0.35$ is the upper limit for the solid phase. In the Fig. 1 (a) case, $\chi$ is equal to 0.1. The value of $\langle (\Delta x)^2 \rangle \simeq 0.15$ may be the limit of the solid phase of the complex plasmas if it has the similar property to the usual crystals but it is not clear at present.

By using Eq. (6), the PDF can be calculated and is shown in Fig. 1 (b). The vertical and horizontal axes are the PDF value and the distance normalized by the nearest neighbor inter-particle distance, respectively. From Fig. 1 (b), it is found that the PDF clearly shows the solid phase since the peaks being far from the origin exist.

The particle displacement is expressed like Eq. (7) but is the two-dimensional function. Here, we assume that the force working between the particles is expressed as $kr$. This is the same as a spring or a harmonic oscillator. Average energy can be calculated in this case, that is,

$$\bar{E} = \int_0^{\infty} \frac{1}{2} k r^2 \frac{1}{2 \pi \sigma^2} \exp \left( -\frac{r^2}{2 \sigma^2} \right) dx = k \sigma^2.$$  

On the other hand, when the velocity distribution function (VDF) is expressed by the two-dimensional Maxwellian, the average energy is known as $E = k_B T$. In the case of the three dimensions, the average energy is $\bar{E} = \frac{3}{2} k_B T$. By comparing these energies, we obtain the relationship of

$$k \sigma^2 = k_B T.$$
where \( k_B \) is the Boltzmann constant. Therefore, the temperature is obtained from this equation if the spring constant is known. If the displacement is regarded as the simple oscillation like \( x = A \exp(i\omega t) \), the oscillation frequency \( \omega \) is described as \( \sqrt{\frac{k}{m}} \). Thus the spring constant is expressed as

\[
k = m \omega^2.
\]  

(12)

By substituting Eq. (12) to Eq. (11), we obtain the relationship of

\[
k_B T = m \omega^2 \sigma^2 \equiv m \omega^2 \langle (\Delta x)^2 \rangle.
\]  

(13)

Therefore, the temperature is obtained from the MSD and the oscillation frequency. This is the same expression of the harmonic oscillation. This means that this method is applicable to only the particle temperature measurements in the solid phase. To obtain the liquid phase temperature, it will be necessary to describe Eq. (13) in another expression. To rewrite Eq. (13) for the liquid phase, further investigation on the particle oscillation in the liquid phase is needed.

Please pay attention that our model is basically applicable to any Coulomb crystals. Its accuracy, however, may be affected by the crystal quality, especially dislocation density.

It is also difficult to obtain the temperature by applying the conventional VDF method to the liquid phase since drift influence must be taken into account. The VDF should be expressed as the shifted Maxwellian at least like,

\[
f(v_x) = n_0 \frac{m}{2\pi k_B T} \exp\left\{-\frac{1}{2} \frac{m(v_x - v_0)^2}{k_B T}\right\},
\]  

(14)

in the one dimension. In this case, the averaged kinetic energy is equal to

\[
\bar{\tau} = \frac{1}{2} k T + \frac{1}{2} m v_0^2.
\]  

(15)

This indicates that drift influence must be eliminated to obtain the particle temperature though it is not easy. The particle temperature measurement in the liquid phase needs further investigations in both methods. Thus we focus on the temperature in the solid phase in this paper.

3. Experimental apparatus

The inside of the apparatus is shown in Fig. 2 (a). There are two RF electrodes of 10 cm in diameter. The distance between the electrodes is 10 cm. Therefore, the coupling between the electrodes is not strong. Each electrode has two RF feeders so that the RF current can flows along the electrode surface. The electrode is flat to achieve flatter potential profiles near the electrode. The nominal diameter of the vacuum chamber is 16 inches. This also helps to keep the flatter potential profile near the edge of the electrode. Figure 2 (b) shows the generated helium plasma without particles. It is shown that the bright area exist near the electrodes. In the region between the electrode and the bright are, there are darker regions. The particles are levitated near the boundary of the bright and dark regions.

One Langmuir single probe is set though it is not shown in this figure. Two CCD cameras are also set. One is set for the horizontal view and the other for top view. The view line of the top view, however, is slanted since the top electrode is an obstacle for the observation.

Intensity of the scattered light from the particles strongly depends on the viewing angle due to the Mie scattering. Therefore, we use macro-lenses for 35 mm cameras to obtain brighter
Figure 2. Inside of experimental apparatus; (a) main parts for plasma generation, dimensions and coordinates and (b) snapshot of RF plasmas. There are two RF electrodes. Each electrode is flat and has two RF feeders. The nominal diameter of the vacuum chamber is 16 inches.

Figure 3. Typical observation result of Coulomb crystal from top view camera on ground. The bright points are the particles of 5 \( \mu \text{m} \) in diameter. The shadow at the upper-left region is the upper electrode. The grids are the shallow ditches made on the back surface, which are used for a scale.

field of views. The magnification in such the lenses is sensitive to the distance between the focal plane and the particles. In addition, images must be corrected to the orthogonal coordinates since those obtained from the top view is slanted. Therefore, We use a specially made glass as a scale. This glass is optically flat (\( \lambda/4 \)) on both surfaces and shallow grids at 5 mm intervals are made on the downward surface touching the lower electrode surface.

The typical observation result of the Coulomb crystal by using this apparatus is shown in Fig. 3. The helium gas pressure and RF oscillator power is set to 0.136 Torr (18 Pa) and 70 W, respectively. The bright points are the particles of 5 \( \mu \text{m} \) in diameter. The shadow at the upper-left region is the upper electrode. The shallow grids are also shown in this picture. It is confirmed that the image is slanted due to the slanted viewing angle of the top view camera. The corrected image is shown in the next section.
4. Results and Discussions

As mentioned previously, the top view data is slanted as shown in Fig. 3. Hence, the data is corrected to the orthogonal coordinates as shown in Fig. 4 (a). This image is obtained by rotating Fig. 3, skewing it and magnifying it vertically. It is confirmed that the grids on the opposite side of the glass is corrected to the rectangular. The shadow of upper electrode is covered by a white sector, which is the same as Fig. 4 (b). Figure 4 (b) is the Delaunay diagram of Fig. 3 (a). It is confirmed that there are some area with highly regular lattices, while there are some dislocations and point defects. Since the highly regular area is a single grain, such the area is suitable for calculating the PDF. From these figures, it is found that the elementary lattice is an equilateral triangle. This is the same structure as that shown in Fig. 1.

The two-dimensional PDF using Eq. (6) is shown in Fig. 5. The solid, dashed and dot-dashed lines represent the experimentally obtained PDF and calculated ones in cases of $\sigma = 0.1$ and 0.15, respectively. In this case, the data in the case of RF power of 100 W and helium gas pressure of 0.136 Torr (18 Pa) are used since more power makes an inter-particle distance shorter. From comparison between the experimental result and the calculations, it is found that the first peak of the experimental PDF ($PDF_{exp}$) agrees well with the calculated PDF ($PDF_{cal}$) in the case of the deviation $\sigma$ of 0.1. The $PDF_{exp}$ apart from the origin, however, does not agree with the $PDF_{cal}$ in the case of $\sigma = 0.1$ but with the $PDF_{cal}$ in the case of 0.15, though the peak position of the $PDF_{exp}$ is well reproduced by these calculations. The good reproducibility of the peak positions can be understood from the coordination number shown in Fig. 6. This figure clearly shows the coordination number of 6.2. This number indicates hexagonal structure.

Although the $\sigma$ of 0.15 may be almost the limit of the solid phase from the Lindemann’s law, the Coulomb crystal seems to still keep the solid phase since the particles do not freely move around but stay and oscillate. This suggests that the limit value of $\chi$ in Eq. (9) may differ from that of actual solids consisting of usual atoms.

If the defects exist, the displacement from the lattice point occurs due to the distortion. Such the displacement is accumulated as the distance increases. Hence, the deviation of $\sigma = 0.1$ at the first peak is considered to indicate the temperature effect in this paper. To calculate the temperature, the oscillation frequency is required. Therefore, the particle trajectories are traced. The typical trajectory of the particle is shown in Fig. 7. In Fig. 7 (a), the horizontal
Figure 5. Comparison of experimentally obtained PDF with calculated ones. Solid, dashed and dot-dashed lines represent an experimental result, calculated results in cases of $\sigma = 0.1$ and 0.15 in Eq. 7, respectively.

Figure 6. Coordination number obtained by integrating PDF. The coordination number is 6.2. This result indicates hexagonal lattice.

and vertical lines are the relative x and y coordinates in the observation area, respectively. The closed circles represent the particle positions, which vary over time. The positions are discrete due to the spatial resolution of the CCD camera. The time evolution of the position is shown in Fig. 7 (b). The offset and the trend of the position is already eliminated. It seems that very low and high frequencies exist. The former one is caused by the particle drift, which includes circular motion. The latter one should relate to the particle oscillation. Since it is difficult to obtain the oscillation frequency directly from Fig. 7 (b), we carry out the Fourier transform to find out the oscillation frequency. The amplitude spectrum is shown in Fig. 8. There are three Fourier transform results in this figure. Thin solid, dashed and dot-dashed lines correspond to these results. By considering the harmonic oscillation, the peak frequency being common throughout these data should exist. The bold dashed lines are located at 0.64, 0.88, 1.29 and 1.64 Hz. The frequencies of 1.29 and 1.64 Hz may be the second harmonics of 0.64 and 0.88 Hz. Since several common peaks exist, Eq. (13) is rewritten as

$$k_B T = m \left\langle (\Delta x)^2 \right\rangle \sum_n \omega_n^2.$$

(16)
Figure 7. Particle trajectory and time evolution of particle displacement; (a) single particle trajectory and (b) time evolution of the displacement of the particle (a). The closed circles in (a) represent the particle position. The position is obtained as discrete data due to the spatial resolution of the CCD camera. The offset and trend in (b) are corrected.

From $\sigma = 0.1$ and the nearest neighbor distance is 0.4 mm, the actual value of $\sigma$ is equal to 0.04 mm. Thus the MSD $\langle (\Delta x)^2 \rangle$ is equal to $1.6 \times 10^{-9}$ m$^2$. The mass of the particle is about $1.31 \times 10^{-13}$ kg from the diameter of 5 $\mu$m and density of 2000 kg/m$^3$. The particle temperature is obtained by substituting these parameters to Eq. (16) as

$$k_B T = 0.29 \ [eV].$$  \hspace{1cm} (17)

To compare this result with the temperature obtained from the VDF, the VDF is obtained and shown in Fig. 9. The open circles are the experimental data. The solid and dashed lines are the Maxwellian. The solid one represents an intermediate line, while the dashed ones the possible minimum and maximum ones. From Fig. 9, it is found that the average velocity is about $v = 1.0 \pm 0.15$ mm/s. This velocity corresponds to the temperature of $E = 0.42 \pm 0.12$ eV. This is about 40% higher than the temperature estimated from the PDF. It is found, however, that non-zero VDF data is located at a few velocity points, namely, the low resolution in velocity space. This is mainly caused by the spatial resolution of the CCD camera as mentioned in section 1. As a result, the temperature from the VDF has an estimation error of 30%. The lowest possible temperature from the VDF is 0.30 eV. This is consistent with the temperature from the PDF.

A weak point of the VDF method is to satisfy both the spacial (velocity) resolution and statistical reliability simultaneously. On the other hand, a weak point of the PDF method is to require the Coulomb crystal with low defect density and an observation duration being long enough to get high frequency resolution of the Fourier transform. Fortunately, these weak points are different from each other. Therefore, both methods can compensate each other and thus the more precise particle temperature measurement will be achieved.

5. Conclusions
The particle temperature in complex plasmas is sometimes regarded to be around the room temperature. The data from our apparatus, however, indicate that the temperature is always higher than that from the VDF. The particle temperature affects some important scientific issues in the complex plasmas such as the critical point and the power and particle balances.
Therefore, we establish another method of the temperature measurement by using the PDF. First, the MSD is determined by comparing the PDF$_{exp}$ with the PDF$_{cal}$. Second, the particle oscillation frequency is determined from the time evolution of the particle coordinates. Then the temperature is estimated by using the MSD and the frequency. It is found that the estimated temperature of 0.29 eV is consistent with that by the VDF method. Since each method has different features and weak points, combination of both methods will lead to compensation of each other’s weak points and the more precise temperature measurement.

Although two or more cameras are needed in the VDF method to get high resolution and large area observations, only one camera is basically needed in the PDF method. Therefore, the PDF method may be more suitable for an apparatus for the microgravity experiment because allowable space and weight are always severe.

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