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Particle ordered structures in a strongly coupled classical thermal plasma

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An ordered structure of macroscopic particles has been experimentally observed in a classical neutral thermal plasma using a laser time-of-flight system. The strongly coupled thermal plasma consists of CeO_2 particles and electrons emitted by the latter under atmospheric pressure and temperature of 1700 K. The particles are charged positively and suspended in a plasma flow. Their charge is about $10^4 e$ and the calculated value of a Coulomb coupling parameter γ_p is >120, which corresponds to a strongly coupled plasma. [S1063-651X(96)51409-1]

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A thermal dusty plasma is a low-temperature plasma containing dust particles in a liquid or solid state. When we introduce dust particles into thermal plasma, they will become charged by collecting electrons and ions, as they do in low-pressure discharges, but also by emitting electrons. The latter process can lead to a positive electric charge, unlike the negative charges in previous low-pressure discharge experiments [1–4]. In the extreme case, particles placed in a nonionized gas completely determine the electrophysical properties of the plasma. These effects were observed in early experiments [5,6] dealing with the plasma in hydrocarbon flames. Since then, the interest in the field of thermophysics and electrophysics of low-temperature thermal plasmas has been sustained owing to the great number of possible applications. These include studies of the properties of rocket-fuel combustion products, the technology of plasma sputtering, and plasma treatment of materials [7].

The thermal plasma under study is at a low temperature, with electrons, ions, and gas all having temperature $T_g = 1700$–2200 K, and at a moderate density, with electron densities in the range $n_e = 10^9$–10^{12} cm^{-3}. It is made from a hot atmospheric-pressure neutral gas. In contrast, the much used rf discharge is nonthermal and made from room-temperature (300 K) gas under low-pressure (1 mbar) conditions.

The thermodynamics of dusty plasma is described by the Coulomb coupling parameter, $\gamma_p = Z_p^2 e^2 / \langle r \rangle k T_g$, which is provided by the ratio between the potential Coulomb energy and the mean thermal energy of the particles. Here Z_p is the particle charge, $\langle r \rangle = (4\pi n_p/3)^{-1/3}$ is the mean interparticle distance, and n_p is the particle density. A plasma with coupling constant γ_p greater than unity may be called a strongly coupled plasma [8]. The strongly coupled dusty plasma exhibits interesting phenomena such as the formation of a more ordered (liquid or solid) structure [8–11]. Recently it was found in laboratory rf plasmas that negatively charged particles tend to self-organize in ordered structures. In a typical experiment, the dust particles (their charge is $\sim 10^4 e$) are embedded in the sheath region where the balance between the gravitational and electrostatic forces is established [1–4]. The plasma sheath, however, where dust particles are trapped, is dominated by ion space charge and is non-neutral [4]. Previously an experimental realization of the crystal-like structures has been already achieved in similar non-neutral systems of macroscopic charged particles [12] and atomic ions [13] in different type of traps.

For a theoretical description of the strongly coupled plasma a one-component plasma (OCP) model and the Yukawa model are usually considered. The classical one-component plasma is an idealized system of ions immersed in a uniform background of neutralizing charges such that the whole system is electrically neutral [8]. The charged particles interact via the Coulomb potential $Z_p^2 e/\langle r \rangle$.

In the Yukawa model shielding effects by the background charge are taken into account, which changes the interaction potential to the Debye-Huckel type: $Z_p^2 e \exp(-\langle r \rangle / r_D)/\langle r \rangle$ [11,14]. Here $r_D = [kT_g / 4\pi e^2 (n_e + n_i)]^{1/2}$ is the Debye shielding length and n_i is the ion density. Then γ_p becomes a function of the screening and the quantity $\Gamma_s = Z_p^2 e^2 \exp(-\langle r \rangle / r_D)/\langle r \rangle k T_g$ is introduced.

The published works on the liquidlike and solidlike structures so far have been restricted to observing the ordered structures for finite non-neutral plasmas consisting of a hundred to a few thousand charged particles [1–4,12,13]. The potential of particle interaction may be quite different in the non-neutral and classical neutral plasmas. For finite plasmas the boundary conditions are predicted to have a significant effect on the plasma state [15]. For example, under conditions of a spherical trap potential, the particle cloud will separate into concentric spherical shells [13]. Instead of a sharp phase transition, the system is evolved gradually from a liquid state characterized by short-range order (liquidlike) to an intermediate state (liquidlike and solidlike), and ultimately to an overall solidlike state [16]. In contrast, the infinite OCP is predicted to exhibit liquidlike behavior for $\gamma_p > 4$ and have a liquid-solid phase transition at $\gamma_c \approx 170$ [8]. The theoretical predictions of Ikezi [9] and Farouki and Hamaguchi [11] also deal with the infinite system of particles interacting via the Yukawa potential.

We present here an experimental study of the formation of a macroscopic ordered structure in a laminar spray of weakly ionized thermal dusty plasma under atmospheric pressure, temperatures of 1700–2200 K, and particle densities up to 10^7 cm^{-3}. This is the classical neutral and extended thermal plasma that is not confined in a trap. The plasma volume and the number of particles being investigated are about of 10 cm^3 and 10^8, respectively. The effect of boundary conditions on the plasma state may be neglected and, in consequence of this, the conditions of the structure formation are close to that in the infinite plasmas.

The experimental facility incorporated the plasma device and the diagnostic instrumentation for the determination of particle and gas parameters [17]. The dusty plasma device uses as the basic plasma source a two-flame Meeker burner with combustion gases seeded with dust particles. The laminar diffusion flame design was used to support a premixed propane-air flat flame and provide a uniform exit profile of the plasma parameters (temperature, velocity, and plasma density). To shield the flame from entrained room air, a central region, 25 mm in diameter, of the burner surface was surrounded by a shroud of combustion gases flowing through an annular area with inner and outer diameters of 25 and 50 mm, respectively. The combustion gas spray velocity V_g was varied from 2 m/s to 3 m/s. In the normal operation of the Meeker burner the plasma density is generally in the range 10^9–10^{11} cm^{-3}, with equal ion and electron temperatures, $T_i = T_e = T_g = 1700$–2200 K. The spectroradiometric measurements of the particles' temperature made on the basis of the technique proposed in [18] show that their temperature is close to the gas temperature $(T_p \cong T_g)$. The combustion gas pressure is 1 atm.

The dust particles were slightly impure and contained sodium and potassium. As a result, the spectra measurements revealed that a plasma spray of particles contains sodium and potassium atoms, which have a low ionization potential.

For studying the collective phenomena and self-organization in the plasma system a knowledge both of the charge on the particles and of the plasma parameters is necessary. An important feature of this plasma device is that it provides a dusty plasma of large enough dimensions (plasma column of 25 mm diameter and 70 mm long) that a variety of plasma experiments may be conducted. Therefore, we are able to make a number of measurements of plasma parameters such as the electron and ion number densities, plasma temperature, and the diameter and number density of the particles.

The local density n_i of positive ions was determined by an electrical probe method [19]. The random error in n_i results in an uncertainty of 20%. To define the electron density n_e, we employ a method based on measurement of the current I and electric field E in the plasma. The equation $j = \sigma E$ is used to obtain the plasma conductivity $\sigma = n_e e \mu_e$ and finally n_e itself [19]. Here j is the current density and μ_e is the electron mobility. The uncertainty in the electron density n_e did not exceed 30%. The measurements of the gas temperature and the densities of sodium and potassium atoms were carried out with the aid of the generalized line-reversion and full-absorption techniques, correspondingly [17].

The original laser method is used for characterization of the mean diameter and density of dust particles in the plasma flow. The method is based on the measurements of transmittance (extinction of a light beam through a dispersed medium) at small scattering angles. This technique is intended for the determination of mean diameter, density, and refractive index of particles in the 0.5–15 μm range [20].

We have observed the macroscopic ordered structure in the plasma spray employing a laser time-of-flight system (see Fig. 1). The blue line ($\lambda = 0.488$ μm) of an Ar^+-ion laser was used as the light source. It was focused near the center line of the burner. The waist diameter at the focal point of the focusing lens was less than 10 μm. The receiving optics observe the measurement volume at a right angle to the laser beam and consist of a receiving objective which images the measurement volume onto the entrance slit of the monochromator. The effective length of the measurement volume is further shortened to approximately 15 μm by the slit. The current pulses emitted by the photomultiplier upon observation of particles were converted to voltage pulses and were transferred to a computer for the subsequent processing. Using our original time series data, we can compute the radial pair correlation function $g(r)$, which represents the probability of finding another particle at a distance $r = V_p t$ away from a given particle [8]. Here V_p is the mean particle velocity, t is the time coordinate, and $V_p \approx V_g$ for micrometer-sized particles.

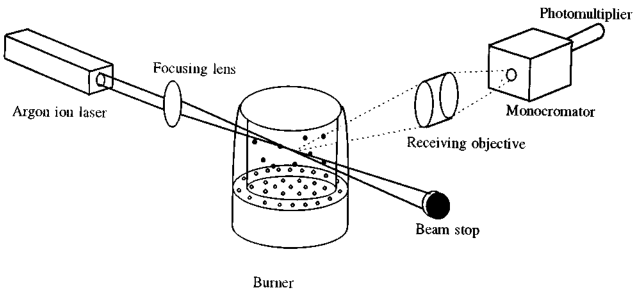

FIG. 1. Schematic of the laser time-of-flight system.

Two types of chemically inert dust in the weakly ionized thermal plasmas were studied in our experiments. The basic constituents of one type were Al_2O_3 particles, electrons, and singly charged Na^+ and K^+ ions, and the other were formed from CeO_2 particles, Na^+ ions, and electrons.

The plasma diagnostics were carried out near the centerline of the plasma column for locations h, ranging from 25 to 40 mm above the burner surface. To study the formation of ordered structures, the plasma measurements were made for different plasma temperatures and electron, ion, and particle densities in the propane-air flame. The plasma temperature was varied by changing the propane-air equivalence ratio ϕ over the range 0.95–1.47. Thus, the Debye length, interparticle distance, and particle charge could also be changed. The particle structure measurements were compared with a random particle distribution obtained at room temperature when only air was supplied to the burner producing the aerosol flow. This simulates a dusty plasma in its ''gas phase.''

The density n_p of CeO_2 particles was varied through the range $(0.25$–$5.0) \times 10^7$ cm^{-3} and the plasma temperature T_g through the range 1700–2200 K. In consequence of this the ion density n_i varied between 0.4×10^{10} and 4.0×10^{10} cm^{-3}, and the electron density n_e ranged from 2.5×10^{10} to 8.0×10^{10} cm^{-3}. The measured mean diameter D_p of particles was 0.8 μm; the rms deviation from nominal size did not exceed 15%.

Based on these data and quasineutrality of the plasma $Z_p n_p + n_i = n_e$, the CeO_2 particles are charged positively and carry about 10^3 electron charges, to within a factor of 2. This observed particle charge can be accounted for by the thermionic emission of electrons from the hot CeO_2 particles

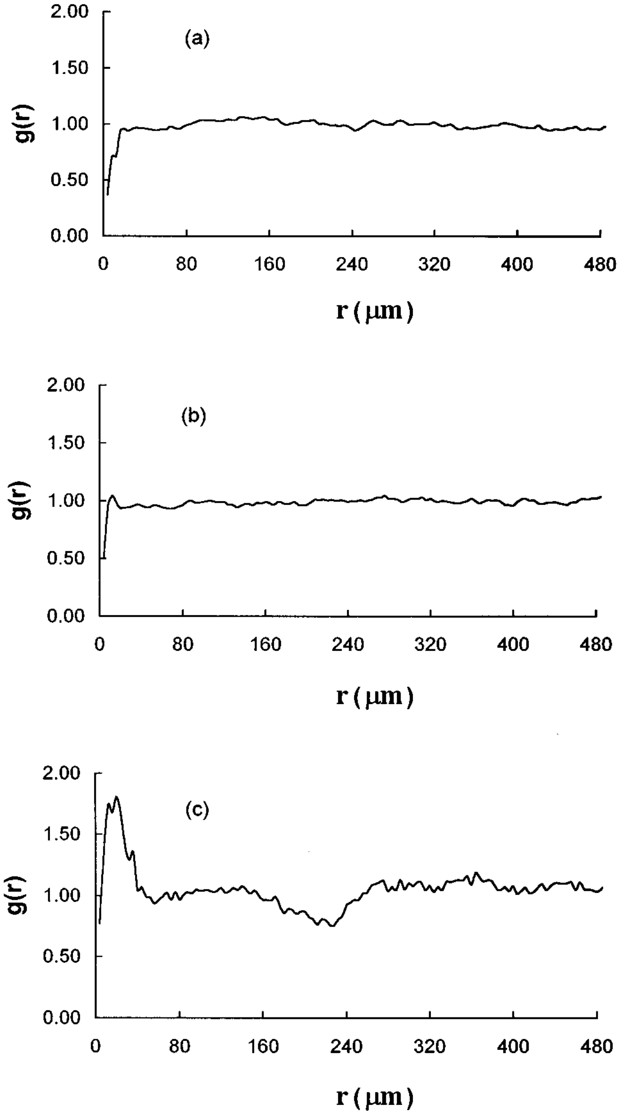

FIG. 2. Pair correlation function $g(r)$ for spray of CeO_2 particles ($Z_p=500$): (a) at room temperature $T_g \cong 300$ K and $\gamma_p=0$ ($Z_p=0$); (b) at plasma temperature $T_g=2170$ K and $\gamma_p=40$ ($\Gamma_s=1$); (c) at plasma temperature $T_g=1700$ K and $\gamma_p=120$ ($\Gamma_s=40$).

[7,10], which have a low electron work function (~2.75 eV). In the analysis of the data given below, the lower limit ($Z_p \approx 500e$) of the particle-charge range is used.

Typical pair correlation functions $g(r)$ for a spray of CeO_2 particles at room-temperature conditions ($T_g \cong 300$ K) and at plasma-temperature conditions ($T_g=2170$ and 1700 K) are shown in Fig. 2. One can see that the pair correlation function computed at a plasma temperature $T_g=2170$ K and a particle number density $n_p=2.0\times10^6$ cm^{-3} are very similar to those observed at room temperature (random gas-like distribution) as shown in Figs. 2(a) and 2(b). Therefore the plasma is weakly coupled and does not exhibit the formation of ordered structure. This fact is also verified by plasma diagnostics. From our optical and probe measurements we obtain that the mean interparticle distance ($\langle r\rangle=50$ μm) is approximately four times the Debye length ($r_D=14$ μm) and that the Coulomb coupling parameter γ_p is about 40. The estimated value of Γ_s is about 1.

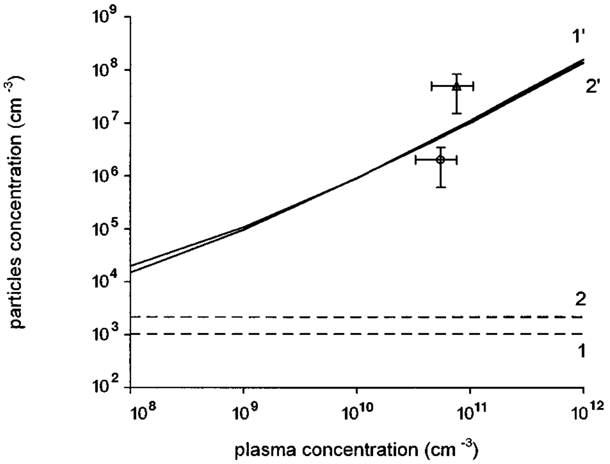

FIG. 3. The range of plasma density (n_e+n_i) and particle density n_p in which an ordered structure is formed when $Z_p=500$. The curves 1 (1′) and 2 (2′) correspond to $\gamma_p=4$ ($\Gamma_s=4$) at $T_g=1700$ and 2200 K, respectively: $\gamma_p=40$ ($\Gamma_s=1$) (○) and $\gamma_p=120$ ($\Gamma_s=40$) (△) for the plasma with CeO_2 particles.

Figure 3 shows the range of n_e and n_p where ordered structure takes place. The theoretical boundaries of the OCP and Yukawa model are indicated. The short-range order condition is satisfied in the region above curves 1 and 2 (OCP model) and curves 1′ and 2′ (Yukawa model). The boundary curves 1 (1′) ($T_g=1700$ K) and 2 (2′) ($T_g=2200$ K) correspond to $\gamma_p=4$ ($\Gamma_s=4$). The Yukawa model predicts higher values of γ_p for the observed interparticle distance $\langle r\rangle$ and the Debye length r_D. The experimental data point (circle) lies between curves 1 (2) and 1′ (2′). The appropriate pair correlation function $g(r)$ is shown in Fig. 2(b). It can be seen that the state of the strongly coupled plasma is found with corresponding values of $\Gamma_s=4$ and $\gamma_p=160$. This value of the coupling parameter γ_p is approximately 40 times larger than the critical value for the OCP model. By this means the experimental data are in accordance with the Yukawa model.

In the case of the lower plasma temperature ($T_g=1700$ K) and particle number density $n_p=5.0\times10^7$ cm^{-3}, Fig. 2(c) shows the pair correlation function $g(r)$, in which the distinctive short-range order of a liquid system is apparent. Under these plasma conditions the ion density ($n_i=4.2\times10^9$ cm^{-3}) is less than the electron density ($n_e=7.2\times10^{10}$ cm^{-3}). Thus the plasma consists of particles and electrons emitted by the latter. The particle charge Z_p is determined by charge balance: $Z_p n_p=n_e$ in this case ($n_i \ll n_e$). The calculated values of γ_p and Γ_s are about 120 and 40, respectively. That corresponds to a strongly coupled system of positively charged particles and electrons. This is a classical one-component system of particles immersed in a background of neutralizing electron gas. The ordered structures in such plasmas have never been observed experimentally. According to the criteria [10] the particles form an ordered structure, which is in agreement with the diagram of plasma states as shown by the triangle in Fig. 3. The corresponding pair correlation function is presented in Fig. 2(c). The experimental point lies above the theoretical lines of the Yukawa model.

In the above calculations of the coupling parameter it is supposed that the dust particles attain a kinetic energy close to the combustion gas temperature. It is possible to make approximate estimates of the thermalization time τ_T for particles from relation $D \approx V_T^2 \tau_T \approx (kT_g/M_p)\tau_T$, where D is the particle diffusion coefficient, V_T is the particle thermal velocity, and M_p is the particle mass. To take into account the effect of the high collision frequency with the neutral gas (the gas is at atmospheric pressure), an empirical interpolation formula [21] is commonly employed:

$$D = (kT_g/6\pi\eta R_p)\{1+(l/R_p) \times [1.257+0.400\ \exp(-1.10R_p/l)]\}, \quad (1)$$

where l is the mean-free path and η is the gas velocity. This relation is true for continuum, free molecular, and transition regimes. So, the time τ_T is defined by

$$\tau_T \approx (2\rho_p R_p^2/9\eta)\{1+(l/R_p) \times [1.257+0.400\ \exp(-1.10R_p/l)]\}. \quad (2)$$

Here ρ_p is the mass density of individual particles. Under typical plasma conditions $T_g = 2000$ K, $R_p = 0.4$ μm, $l \approx 0.4$ μm, $\rho_p = 7.3$ g/cm^{-3}, and $\eta = 7.0\times10^{-5}$ Pa s; the time τ_T is estimated to be ~ 10 μs. This value is much less than the characteristic time of plasma life $\tau_f = h/V_p \cong 20$ ms ($h \cong 40$ mm, $V_p \cong 2$ m/s).

We observed the short-range structure only at high particle densities (up to $\sim 10^7$ cm^{-3}). Decreasing the density of CeO_2 particles increases the mean interparticle distance and causes a reduction in the Coulomb energy. The spatially ordered structure no longer holds, as is seen in Fig. 3(b) for $n_p = 2.0\times10^6$ cm^{-3}.

The plasma with Al_2O_3 particles was studied at temperatures in the range $T_g = 1900$–2200 K. It is worth noting that the number density of Na^+ and K^+ ions in the flame with Al_2O_3 particles is greater than the density of ions in the plasma spray with CeO_2 particles by a factor of 10. The measured densities of ions, electrons, and particles lie in the ranges $(0.35$–$12)\times10^{10}$ cm^{-3}, $(0.85$–$18)\times10^{10}$ cm^{-3}, and $(0.7$–$1.0)\times10^6$ cm^{-3}, respectively, according to our diagnostic measurements. The mean size of particles was about 1.5 μm.

Due to the larger numbers of alkali ions and electrons, the Debye screening reduces the Coulomb interaction. For example, taking $T_g = 2035$ K, $n_i = 8.6\times10^{10}$ cm^{-3}, $n_e = 1.3\times10^{11}$ cm^{-3}, and $n_p = 1.0\times10^6$ cm^{-3}, we obtain the values $r_D = 6.5$ μm and $\langle r\rangle = 60$ μm. Since $\langle r\rangle \cong 9r_D$, the particles are significantly shielded from each other and do not form a space-ordered structure.

In conclusion, we have experimentally observed an ordered structure of particles in the classical neutral thermal plasma using a laser time-of-flight system. The strongly coupled thermal plasma consists of CeO_2 particles and electrons emitted by the latter. The particles are charged positively and suspended in a plasma flow. Analysis of the pair correlation function reveals an ordered structure, which is consistent with the large value of the Coulomb coupling parameter obtained from plasma measurements. The conditions of the formation of ordered structures are close to that in the infinite plasmas of the Yukawa type. Upon decreasing the particle density and increasing the alkali-metal-ion density we observed a chaotic distribution of particles in space. The diagnostic instruments allowed us to measure the parameters of thermal dusty plasmas and to study the strongly coupled plasma systems.

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