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EXPERIMENTAL OBSERVATION OF COULOMB ORDERED STRUCTURE IN SPRAY OF THERMAL DUSTY PLASMAS

V.E.Fortov, A.P.Nefedov, O.F.Petrov, A.A.Samarian, A.V.Chernyschev, A.M.Lipaev

High Energy Density Research Center RAS

127412 Moscow, Russia

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A macroscopic Coulomb ordered structure of polydisperse CeO_2 particles in a laminar spray of weakly ionized thermal plasma under atmospheric pressure and temperature of about 1700 K has been experimentally observed. Diagnostic instruments allowed one to measure plasma parameters. The particles are charged positively and have of about 10^3 electron charges. The calculated values of Coulomb coupling parameter γ_p is > 120 that corresponds to a strongly coupled plasma.

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A dusty plasma can be defined as ensemble of charged dust particles immersed in a plasma. A plasma with coupling constant γ_p, defined as the ratio of the average Coulomb energy to the average thermal energy, greater that unity may be called strongly coupled plasma [1]. The strongly coupled dusty plasma exhibits interesting phenomena such as the formation of a more ordered (liquid or solid) structures [1]. Recently it was found in laboratory rf plasmas that negatively charged particles tend to self-organize in ordered structures as theoretically predicted by Ikezi [2]. 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 [3, 4].

Regardless of recent experimental progress in the study of Coulomb solids in the above mentioned systems, the ordered structure in thermal dusty plasmas has never been observed experimentally. The electron density n_e in a thermal plasma consisting of ionized gas and dusty particles is specified by two processes. One of these processes is the ionization of gas molecules and recombination, and the other is related to thermionic emission from and recombination of electrons with the particles [5]. In the limit case, the thermal plasma can be consisted of only particles with positive charge and free electrons emitted from their surface. The entire range of states, from a Debye plasma to a highly nonideal system of charged particles, is realized in a plasma under standard conditions ($T_g = 1700 - 3000$ K, $n_e = 10^9 - 10^{12}$ cm^{-3}) owing to high values of the particle charges [6]. Here we report an experimental observation of a macroscopic Coulomb ordered structure in a laminar spray of weakly ionized thermal dusty plasma under atmospheric pressure and temperature of about 1700 K.

The experimental facility incorporated the plasma device and the diagnostic means for the determination of particle and gas parameters [7]. Our dusty plasma device uses, as the basic plasma source, a two-flame Meeker burner with combustion gases seeded with dusty 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 conditions (temperature, velocity, and plasma density). To shield the flame from entrained room air, a 25 mm diameter central region of 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 ranged from 3 m/s to 5 m/s. In the normal operation of the Meeker burner the plasma density is generally in the range $10^9 - 10^{11}\,\mathrm{cm}^{-3}$, with equal ion and electron temperatures, $T_i = T_e = T_g = 1700 - 2200$ K. The spectroradiometric measurements of particles temperature made on the basis of the technique proposed in [8] show that their temperature is close to the gas temperature ($T_p \simeq T_g$). The combustion gas pressure is an atmospheric.

The dust particles were slightly impure and contained sodium and potassium. As a result, the spectral measurements revealed that a plasma spray of particles contains sodium and potassium atoms with a low ionization potential. Two types of the weakly ionized thermal plasmas with chemically inert dust have been studied in our experiments. The basic constituents of one were the Al_2O_3 particles, electrons, and singly charged Na^+ and K^+ ions, and the other one were formed from the CeO_2 particles, Na^+ ions and electrons.

For studying the Coulomb ordered structure a reliable knowledge of basic plasma parameters is required. We are able to make a number of measurements the plasma parameters such as electron and ion number densities, plasma temperature, diameter and number density of particles. The local density n_i of positive ions was determined by an electrical probe method [9]. The random error in n_i results in an uncertainty of 20 %. To define the electron density n_e, we employ the method based on the measurement of 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 [10]. 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 densities of sodium and potassium atoms were carried out with the aid of the generalized line reversal and full absorption techniques, correspondingly [7]. The original laser method is used for characterization of the mean diameter and density of dusty particles in a 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 micron range [11].

We have observed the Coulomb ordered structure in the plasma spray employing a laser time-of-flight system (see Fig.1). The blue line ($\lambda = 0.488\,\mu$m) of an Ar^+-ion laser was used as the light source. It was focused near the centerline of the burner. The waist diameter at the focal point of focusing lens was less then 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 monocromator. 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 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 [1]. Here V_p is the mean particles velocity ($V_p \simeq V_g$), and t is the time coordinate.

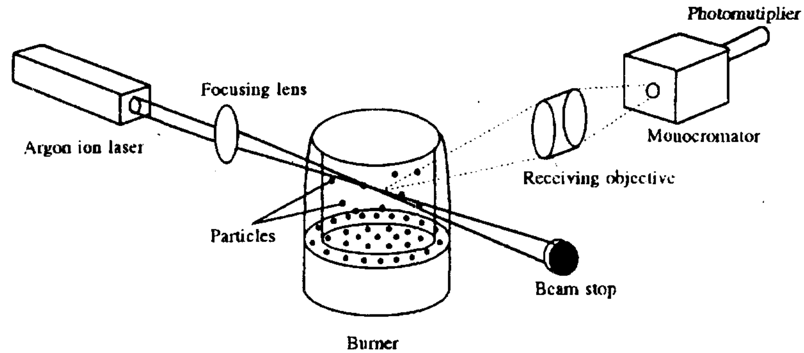
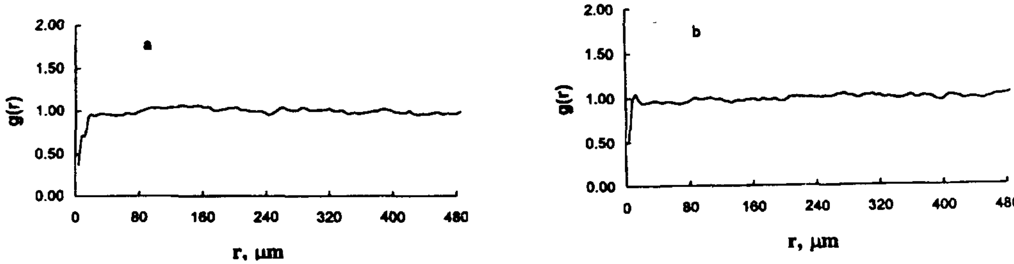


Fig.1. Schematic of the laser time-of-flight system

The plasma diagnosis has been taken for locations, h, ranging from 25 to 40 mm above the burner surface. To study the formation of ordered structure, the plasma measurements were made for different plasma temperatures, electron, ion and particle densities in the propane-air flame. The plasma temperature was varied by changing the equivalence propane-air ratio ϕ in the limits of 0.95-1.47. So, the Debye length, interparticle distance and particle charge could be also changed. The particles structure measurements were compared with a random particles distribution obtained at room temperature when only the air were supplied into burner producing the aerosol flow. This simulates a dusty plasma in its "gas phase".

Typical pair correlation functions g(r) for spray of CeO_2 particles at room temperature conditions ($T_g \simeq 300$ K) and at plasma temperature conditions (T_g=2170 K and T_g=1700 K) are shown in Fig.2. The particle density n_p was varied through range $(0.2 - 6.8) \cdot 10^7\,\text{cm}^{-3}$ and the plasma temperature T_g was changed in the limits of 1700–2200 K. In consequence of this the ion density n_i varies between $0.42 \cdot 10^{10}$ and $4.0 \cdot 10^{10}\,\text{cm}^{-3}$, and the electron density n_e ranges from $2.5 \cdot 10^{10}$ to $7.2 \cdot 10^{10}\,\text{cm}^{-3}$. The measured particles mean diameter $D_p = 2R_p$ was 0.8 μm. 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 have of about 10^3 electron charges with an error of a factor 2. The observed particle charge can be explained by the thermionic emission of electrons from the hot CeO_2 particles [5,6,12,13] of low work function (~ 2.75 eV [14]). In the analysis of the data that given below the lower limit ($Z_p \simeq 500e$) of the particle charges range is used.

One can see that pair correlation function computed at plasma temperature T_g=2170 K and particles number density $n_p = 2.0 \cdot 10^6\,\text{cm}^{-3}$ is very similar to those observed at room temperature (random gaslike distribution). Therefore the plasma is weakly coupled and the formation of ordered structure is not exhibited. This fact is also verified by plasma diagnosis. From our optical and probe measurements we obtain that mean interparticle distance ($< r > =$ 50 μm) is approximately four times the Debye length ($r_D = 14$ μm) and the Coulomb coupling parameter $\gamma_p = Z_p^2 e^2 / < r > kT_g$ is about 40. In order to take the Debye-shielding effect into account, the Coulomb energy is replaced by the shielded Coulomb potential $(Z_p e)\exp(- < r > /r_D)/ < r >$ and the quantity



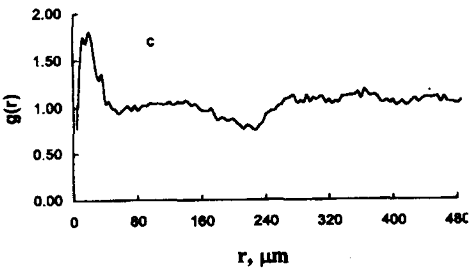


Fig.2. Pair correlation function $g(r)$ for spray of CeO_2 particles: *a*) at room temperature conditions, $T_g \simeq 300$ K; *b*) at plasma temperature T_g=2170 K; *c*) at plasma temperature T_g=1700K

$\Gamma_s = Z_p^2 e^2 \exp(- < r > /r_D)/ < r > kT_g$ is introduced. The estimated value of Γ_s is about 1.

Fig.3 shows a range of n_e and n_p where ordered structure takes place. The boundary curves *1* (T_g=1700 K) and *2* (T_g=2200 K) correspond to $\Gamma_s = 4$. The short-range order condition is satisfied in the region above the curves [6]. The experimental data point (close circle) lies near boundary between the states of strongly and weakly-coupled plasmas.

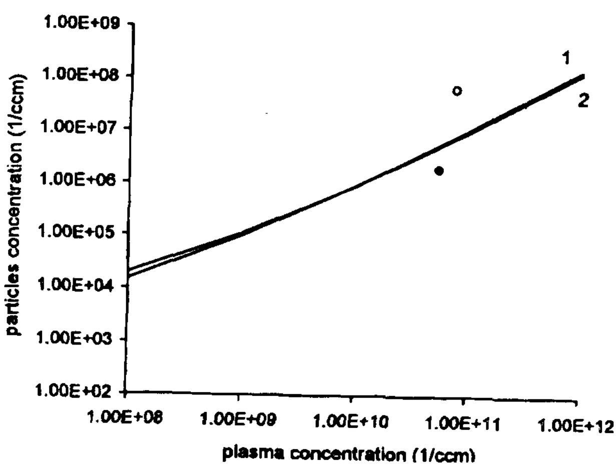


Fig.3. A range of plasma density $n_e + n_i$ and particle density n_p where an ordered structure is formed when $Z_p = 500$. The curves *1* and *2* corresponds to $\Gamma_s = 4$ at T_g=1700 K and 2200 K, respectively: $\Gamma_s = 1$ (•) and Γ_s=40 (o) for the plasma with CeO_2 particles

In the case of lower plasma temperature (T_g=1700 K) Fig.2c shows the pair correlation function g(r), in which the distinctive short-range order of a liquid system is apparent. In these plasma conditions the number density of ions ($n_i = 4.2 \cdot 10^9\,cm^{-3}$) is lesser than that of electrons ($n_e = 7.2 \cdot 10^{10}\,cm^{-3}$), and the particles number density is $6.8 \cdot 10^7\,cm^{-3}$. 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 the strong coupled system of positively charged particles and electrons. According to criteria [2] the particles form an ordered structure which is in agreement with diagrams of plasma states as shown in Fig.3 by open circle.

We observed the short range structure only at high particle densities (up to $\sim 10^7\,\mathrm{cm}^{-3}$). Decreasing the density of CeO_2 particles increases the mean interparticle distance and causes the reduction of Coulomb energy. The space ordered structure no longer holds as seen in Fig.2b for $n_p = 2.0 \cdot 10^6\,\mathrm{cm}^{-3}$.

The plasma with Al_2O_3 particles was studied at the temperatures in the range T_g=1900-2200 K. It is worth noting that the number density of Na^+ and K^+ ions in flame with Al_2O_3 particles is more than density of ions in the plasma spray of CeO_2 particles by a factor of 10. The measured densities of ions, electrons and particles lie in the range $(0.35 - 12) \cdot 10^{10}\,\mathrm{cm}^{-3}, (0.85 - 18) \cdot 10^{10}\,\mathrm{cm}^{-3}$ and $(0.7 - 1.0) \cdot 10^6\,\mathrm{cm}^{-3}$, respectively from our diagnostic measurements. The mean size of particles was about 1.5 μm. Due of to the larger amount of alkali ions and electrons, the Debye screening reduces the Coulomb interaction. For example, taking $T_g = 2035\,\mathrm{K}$, $n_i = 8.6 \cdot 10^{10}\,\mathrm{cm}^{-3}$, $n_e = 1.3 \cdot 10^{11}\,\mathrm{cm}^{-3}$ and $n_p = 1.0 \cdot 10^6\,\mathrm{cm}^{-3}$ yield the values $r_D = 6.5\ \mu$m and $< r > = 60\ \mu$m. Since $< r > \simeq 9 r_D$ the particles are significantly shielded from each other and do not form space-ordered structure.

In conclusion, we have demonstrated the formation of the Coulomb ordered structure in the laminar spray of the strongly coupled thermal dusty plasma using the laser time-of-flight system. Analysis of pair correlation function reveals the ordered structure, which is consistent with a large value of the Coulomb coupling parameter obtained from plasma measurements. Decreasing particles density and increasing alkali ions density we have observed the chaotic positions of particles. The diagnostic instruments allowed one to measure parameters of thermal dusty plasma and to study the strongly coupled plasma system.

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