Crystallization of a dusty plasma in the positive column of a glow discharge

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The formation of macroscopic ordered structures in the standing striations of a stationary glow discharge in Ne is observed. A Coulomb quasicrystal is formed by spherical glass particles with diameters of 50–63 μm and charge $Z_p \sim 7 \cdot 10^5 e$. The interparticle distance is approximately 300 μm. This corresponds to a nonideality parameter $\Gamma \sim 5 \cdot 10^4$, which leads to crystallization in the Yukawa model. The factors leading to the formation of a quasicrystal in the striations are discussed. © *1996 American Institute of Physics.* [S0021-3640(96)00514-2]

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A dusty plasma is a low-temperature plasma containing finely dispersed liquid or solid particles. Such a plasma is sometimes called an aerosol plasma or a plasma with a condensed disperse phase (CDP). The presence of condensed particles can substantially shift the ionizational equilibrium in the plasma and lead to a number of new effects. In recent years, on account of the extensive use of plasma spraying and etching technologies in microelectronics and in thin-film production, great interest has been shown toward the study of the properties of dusty plasmas.[1,2] The presence of particles in a plasma not only results in contamination of the surface of semiconductor elements and thereby a larger number of defective elements, but in addition the plasma is perturbed, often in an unpredictable manner. These deleterious effects cannot be diminished or prevented without an understanding of the processes involved in the formation and growth of condensed particles in a gas-discharge plasma, the mechanism of their transport, and their effect on the properties of the discharge.

Ordinarily, the plasma in a low-pressure rf discharge in a gas is employed for surface treatment.[3] The degree of ionization of such a plasma is low ($\sim 10^{-7}$), the electron energy equals several eV, and the ion energy is close to the thermal energy of the atoms (≈ 0.03 eV). A neutral nonemitting particle in such a plasma is bombarded by all particles present in the plasma, including electrons and ions. It is conventionally assumed that electrons impinging on the surface of a particle are completely absorbed and that ions impinging on the surface of the particle tear out electrons and recombine. As a result of the large difference in the electron and ion masses, the electron flux is several orders of magnitude higher than the ion flux, and a particle will become negatively charged. The negative electrostatic potential appearing on a particle repels electrons and attracts ions.

The charge of the particle continues to change until the electron and ion fluxes on the particle become equal to one another. The typical value of the steady-state charge of a micron-size particle ranges from 10^3 to 10^5 electron charges. Correspondingly, the walls of the discharge chamber and the electrodes acquire a negative potential. Under certain conditions, all this makes it possible to compensate gravity and results in levitation (hovering) of the particles above the bottom electrode or the bottom of the discharge chamber. Methods for confining dusty plasmas in special traps, making it possible to decrease the contamination of the surfaces being treated, are based on this effect.

The pair interaction energy between charged particles is proportional to the product of the charges of these particles. In a dusty plasma, where the particle charge Z_p is large, nonideality can appear in the interparticle interaction much earlier than in the electron–ion subsystem, despite the fact that the particle density is ordinarily low compared with the electron and ion densities. The nonideality parameter Γ of a system of charged particles is usually determined by the ratio of the potential energy of the Coulomb interaction to the kinetic energy of the thermal motion

$$\Gamma = \frac{Z_p^2 e^2}{aT}, \tag{1}$$

where T is the temperature of the particles and

$$a = \left(\frac{3}{4\pi N_p} \right)^{1/3} \tag{2}$$

is the radius of the sphere per particle (Wigner–Seitz radius). If the particle charge Z_p is set equal to 10^3 and the particle density N_p is set equal to 10^5, then Γ is indeed of the order of 10^2, which attests to the strong nonideality of a dusty plasma under these conditions. It is known from the simplest and best-studied model of a one-component plasma that short-range order appears in the system for $\Gamma > 1$ and the one-component plasma crystallizes at $\Gamma \cong 170$.[4]

The conditions for possible crystallization of a dusty plasma were formulated in Ref. 5. However, almost 10 years passed before a dust crystal was observed in an rf discharge plasma. This occurred almost simultaneously in four laboratories: in Taiwan,[6] Germany,[7,8] and Japan.[9] In recent years, ordered structures of macroparticles have been observed in a thermal plasma at a temperature of approximately 1700 K.[10]

In an rf discharge, a plasma crystal forms near the bottom electrode at the boundary of the cathode sheath. The crystal can have various structures with lattice constants of the order of a fraction of a millimeter. This makes it possible to observe the crystal with a virtually unaided eye. Plasma crystals possess a number of unique properties which make them an indispensable tool both in the investigation of the properties of strongly nonideal plasma and in the investigation of the fundamental properties of crystals.

Thus far, all attempts to obtain a plasma crystal in a stationary glow discharge rather than an rf discharge have been unsuccessful. This is apparently attributable to the fact that despite an increase in the electric field intensity near the cathode, the decrease in the particle charge as result of the lower electron density makes it impossible for the electrostatic forces to completely compensate gravity. Only an rf field, which periodically

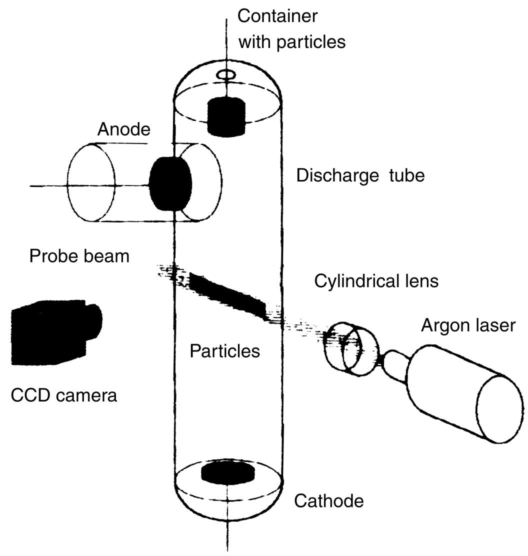

FIG. 1. Diagram of the experimental apparatus.

transfers electrons deep into the cathode sheath, results in a compensation of these forces and levitation of dust particles. Nonetheless, in the present work ordered quasicrystalline structures were observed to form in a stationary glow discharge. However, the crystallization occurs not in the cathode sheath but in standing striations.

A glow discharge was produced in a cylindrical glass gas-discharge tube with cold electrodes. The inner diameter of the tube was 3 cm and the distance between the electrodes was 40 cm. A diagram of the apparatus is displayed in Fig. 1. The discharge current ranged from 1 to 10 mA and the neon pressure ranged from 0.4 to 2 torr. Discharge regimes with natural standing striations existed in this range.

Borosilicate glass ($\rho = 2.3$ g/cm^3) particles in the form of hollow thin-wall spheres 50–63 μm in diameter were introduced into the plasma in the positive column of the discharge. The particle walls were approximately 5 μm thick and the loose-fill density of the particles was close to 0.9 g/cm^3. The particles were placed inside a cylindrical container, whose bottom consisted of a metal grid with a spacing of 100 μm. When the container was shaken, the particles fell and entered the positive column. The particles were visualized by illumination with a probe laser beam in the horizontal or vertical planes. A cylindrical lens shaped the beam from an argon laser into a flat converging beam with a waist 20 mm wide and 150 μm thick at the center of the discharge tube. The horizontal probe beam could be moved over the height of the tube and the vertical probe beam could be moved over both the height and radius of the tube. The reflected light was observed with the aid of a CCD camera at an angle of 45° in the case of the horizontal beam and 90° in the case of the vertical beam and was recorded with a videotape recorder.

When standing or weakly oscillating striations were present in the positive column

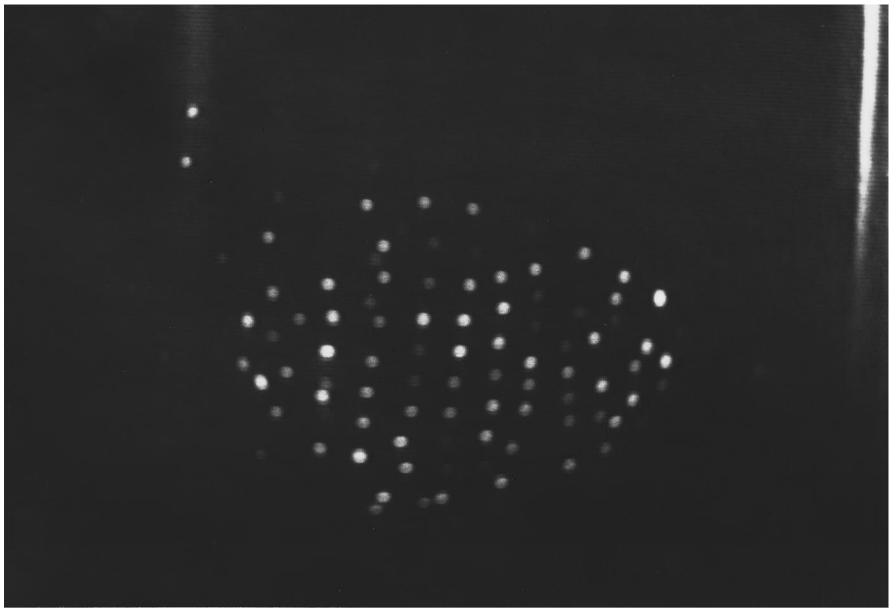

FIG. 2. Photograph of ordered structure in a striation in a vertical plane at a discharge current of 1.1 mA and a neon pressure of 0.4 torr.

of the discharge, charged macroscopic particles, forming ordered quasicrystalline structures whose size and shape depend on the parameters of the discharge, were observed to hover in the glowing parts of the striations. The structure forms as follows: After the container is shaken, the particles overshoot the equilibrium position, ‘‘float upward’’ for several seconds, and align themselves in an ordered structure, which, if the discharge parameters are constant, persists indefinitely. Individual particles can move upwards toward the anode along the periphery of the striations. We note that ordered structures were observed in several neighboring striations simultaneously. As a striation fluctuates, the cloud of particles fluctuates together with the glowing region of the striation. In the case of strong oscillations, individual particles can fall out of the structure. Figures 2 and 3 display examples of the structures observed in the vertical and horizontal planes. Approximately 10 horizontal layers of particles were observed by scanning the horizontal beam in the vertical direction. The layers were separated by 260–320 μm, and the distances between the particles in the horizontal plane equalled 350–600 μm.

The basic properties of striations in low-pressure discharges have been well studied.[11–14] In inert gases under conditions of inelastic electron energy balance, natural periodic structures with a characteristic scale $\lambda_1 \approx \epsilon_1 / eE_0 \cong 4-5$ cm arise (ϵ_1 is the first excitation potential, equal to 16.6 eV for neon, and E_0 is the average electric field over the length of a striation). At the head of a striation—a brightly luminescing region of strong fields which occupies approximately one third of the striation, the electron density increases by an order of magnitude. Then ionization stops, since the field practically

FIG. 3. Photograph of ordered structure in a striation in a horizontal plane at a discharge current of 0.9 mA and a neon pressure of 1.0 torr.

vanishes and electrons are lost on account of diffusion toward the walls. The electron density gradually decreases right up to the head of the next striation. The dark region is several times longer than the glowing region. The fact that the current remains constant along the discharge implies that the field starts to increase as the density decreases, and everything repeats in the next striation. The striations are substantially two-dimensional. At the head of a striation the center–wall potential difference reaches 20–30 V, the change in the potential occuring in a narrow 2–3 mm thick layer near the wall.[13,14] In the strong-field region, a secondary maximum appears in the electron energy distribution function, shifts to higher energies along the striation, and vanishes at the end of the striation, having reached the excitation energy ϵ_1.

At the head of a striation, the charge of the macroscopic particles increases sharply as well as the longitudinal electric field. This is most easily verified by equating the floating potential φ_p of a particle to the wall potential $\varphi_w \cong 30$ V. Since $Z_p \cong \varphi_p R_p / e$ (R_p is the particle radius), we obtain $Z_p \cong 7 \cdot 10^5$. To prevent a particle with mass M_p from falling, the electric force $Z_p e E$ must compensate the gravitational force $M_p g$, whence

$$E_m = \frac{M_p g}{Z_p e}. \qquad (3)$$

The mass of a particle 63 μm in diameter with walls 5 μm thick equals 10^{-7} g. This gives $E_m \cong 88$ V/cm, which is three to four times higher than the fields ordinarily observed at the head of a striation. However, this disparity is easily explained by the separation of the particles according to wall thickness (and therefore according to mass) directly in the discharge: particles with the lowest value of the parameter M_p/Z_p, primarily thin-walled particles, are trapped.

A large value of Z_p leads to a strong interparticle Coulomb repulsion and strong nonideality of the system. For an average interparticle distance $a=300$ μm and temperature $T=300$ K, the nonideality parameter $\Gamma\sim 10^8$. However, it is necessary to take account of the fact that the particles are screened by the plasma electrons and ions, whose density at the head of a striation is $\sim 10^9$ cm^{-3}. The Debye radius $r_D \cong 40$ μm ($a\cong 10r_D$), which attests to the strong screening. The screening effect is taken into account in the Yukawa model,[15] where an interaction potential of the Debye–Hückel type is introduced:

$$\Phi_D(r)=\frac{Z_p e}{r}e^{-r/r_D}. \quad (4)$$

Numerical calculations on the basis of the Yukawa model show that crystallization requires $\Gamma=4.8\cdot 10^4$ with $a/r_D=10$. The characteristic value $\Gamma\sim 5\cdot 10^4$ for our experimental conditions is in good agreement with the results of these calculations. The incomplete regularity of the observed structures is apparently associated with the inadequate monodispersity of the particles with respect to mass and charge. A complex is prevented from falling apart in the radial direction by the presence of a strong radial electric field, reaching values of $\sim 10^2$ V/cm, near the wall. However, inside a complex itself this field is weak and has virtually no effect on the parameters of the ordered structure.

In conclusion, we note that we have observed the formation of ordered structures of charged macroscopic particles of large size and weight in the striations of a low-pressure glow discharge. Confinement of a complex is achieved through a high charge on the particles and the presence of strong axial and radial gradients of the potential at the head of the striations.

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