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PLASMA DUST CRYSTALLIZATION¹J. Goree²University of Iowa
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In a ground-based definition study, a concept for a new type of microgravity experiment is developed. We formed a new state of matter: a crystalline lattice structure of charged micron-size spheres, suspended in a charge-neutral plasma. The plasma is formed by a low-pressure radio-frequency argon discharge. Solid micro-spheres are introduced, and they gain a negative electric charge. They are cooled by molecular drag on the ambient neutral gas. They are detected by laser light scattering and video photography. Laboratory experiments have demonstrated that a two-dimensional non-quantum lattice forms through the Coulomb interaction of these spheres. Microgravity is thought to be required to observe a three-dimensional structure.

INTRODUCTION

According to theory, plasmas can crystallize as they cool. What makes a crystal extraordinary is that virtually every known plasma, astronomical and man-made, is a gas-like fluid, not a solid.

Consider individual charged particles: those with identical charges repel one another and attempt to maintain a separation between themselves. But they cannot move infinitely far apart if they are electrically or magnetically confined in a volume. Nor can they move apart if there is a neutralizing background of the opposite polarity. This is called a one-component plasma (OCP). Such OCPs have been studied in other experiments, as we will review in Section III. We interest ourselves here in a kind of plasma that is closely related to the one-component plasma, but differs in the way that the neutralizing background behaves.

Usually in a plasma the thermal energy is high enough that the plasma behaves like a gas, with random particle velocities and trajectories with long mean-free-paths. This is called a "weakly-coupled plasma," and it is like a gas. By cooling the particles and lowering the thermal energy, according to theory, the charged particles in a plasma will adopt fixed positions with a uniform separation in order to minimize their potential energy. This is a crystal, and it is the extreme case of a "strongly-coupled plasma." It arises from the internal electrostatic interaction between particles and the containment of an external potential well.

The OCP can undergo phase transitions between gaseous, liquid and solid states. These states differ from ordinary states of matter in that the particles are much larger (micron size) than atoms, they are

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spaced more widely (hundreds of microns) and their inter-particle interaction is strictly classical electrodynamics. The thermodynamics is quantified by the density of the grains and by their temperature, which are combined into the dimensionless parameters

$$\Gamma = Q_p^2 / (k_B T \Delta)$$

$$\kappa = \Delta / \lambda_D,$$

where Q_p is the charge on the particles, T is their temperature, Δ is the typical inter-particle spacing, and $\lambda_D \approx (\epsilon_0 k T_i / n_i e^2)^{1/2}$ is the Debye shielding length. Physically, Γ is the ratio of the electrostatic potential energy to the thermal kinetic energy of a grain. By cooling the particles, one increases Γ . We cool the grains by drag on rarefied neutral gas in the plasma.

Monte Carlo simulations predict that, in the absence of additional disruptive forces, the OCP behaves as a:

gas	for $\Gamma < 2$
liquid	for $\Gamma > 2$
solid	for $\Gamma > 178$.

For high values of Γ , the OCP is a solid crystal, sometimes referred to as a "Wigner crystal," which is a non-quantum ordered structure in the cases we will consider.

A plasma containing micron-size grains has several advantages in forming a crystalline lattice: the charge Q_p is typically thousands of electron charges so it is easy to make Γ large, and the individual grains are big enough to be imaged photographically.

One can compute Q_p from plasma theories that were originally developed for charged bodies such as spacecraft and dust particles. [1-2] The sphere collects both electrons and ions, and so it gathers a net current until a steady state is reached where electron and ion currents exactly cancel. This steady state Q_p is negative, since electrons are faster than ions and must be repelled to reduce the electron current to equal the ion current. The negative charge is just like that of an object in a space plasma. Its value is given by $Q_p = C V_f$, where $C = 4\pi\epsilon_0 r_p$ is the capacitance of a sphere of radius r_p , and V_f is its steady-state electric potential.

In our experiment the problem is complicated from that of the simple OCP model because the neutralizing background of electrons and ions is free to respond to the electric potential of the charged grains. As a result, Γ is not by itself a sufficient thermodynamic parameter to describe the phase transitions. Farouki and Hamaguchi [3] recognized this, and introduced κ as the other thermodynamic variable. Together, Γ and κ are a complete set of variables for the thermodynamic system, equivalent to using the temperature and density as the variables. Farouki and Hamaguchi have also developed a Molecular-dynamics simulation of the phase transition from solid to crystalline dusty plasmas. [3,4] They have also introduced the idea that the negatively-charged grains are not purely repulsive, because they are surrounded by a net positively-charged cloud of ions in a Debye sheath, which also interact as the grains are brought within one Debye length of one another. [4]

Based on the OCP theory and the results of Farouki and Hamaguchi, it is expected that room temperature is cool enough for our spheres to crystallize, if they have charges of a thousand electron charges, or more.

We have carried out laboratory experiments that have demonstrated the formation of a 2-dimensional crystal, verifying in part the theories. These lab experiments are described below, and in a forthcoming paper. [5]

APPARATUS DESIGN

Our laboratory definition studies reveal that a design using an RF powered plasma is feasible because it levitates and confines dust particulates for hours. [5] In fact, we chose the RF design because it is known from its commercial use in plasma processing of materials that it easily levitates and confines charged grains. [6] In the laboratory, gravity causes the particulates to be confined in a thin horizontal layer. In a microgravity environment we expect that they will disperse throughout the central region of the plasma. Drop-tower and parabolic flights may be used to test this assumption.

RF discharge operation is understood well. Electric sheaths form at the two electrodes, and their thicknesses oscillate with the applied rf voltage. The sheath edges collide with the electrons in the plasma and accelerate them stochastically to the energies required to sustain the plasma by electron-impact ionization of the neutral gas. RF power is applied continuously to sustain the discharge in a steady state. Ions and dust particles move too slowly to respond to the RF electric fields; instead they respond to a time-average DC electric field that forms naturally. This DC electric field points everywhere outward from the center of the plasma, providing stable confinement of the negatively-charged dust particulates. Dust is confined in the large central region of the plasma, where the field is less than 1 V/cm, small enough not to tear apart a lattice.

The RF design will allow an adjustable RF output power. This will allow a variable plasma density, which is useful for multiple experimental runs to test the density dependence in theories involving Γ and κ . A generator with a maximum output of 10 W should allow plasma densities of $\leq 10^{10} \text{ cm}^{-3}$.

There are two practical difficulties of using an RF discharge that we must evaluate during the laboratory definition study stage. These are thermal gradient forces and ion-drag forces. Either of them could disrupt crystal formation. Thermal gradients within the gas can disrupt the crystal formation due to the "thermophoresis force" [7] of the gas on the particles. During the laboratory experiments we will quantify this effect and arrive at engineering criteria for thermal design of the flight hardware. Ion-drag forces [8] arise from the ions that are streaming from the center of the plasma toward the negatively-biased electrodes. The ions are scattered by Coulomb collisions with the massively charged grains, and they impart some momentum to the grains. Expressions have been developed for ion drag for an isolated grain, [8] but it is unknown how substantial this force will be for ions passing through a cloud of grains, as in our experiment.

RESULTS OF LABORATORY DEFINITION EXPERIMENTS

A macroscopic Coulomb crystal of solid particles in a plasma has been observed in our laboratory definition experiments. Images of a cloud of 7- μm "dust" particles, which are charged and levitated in a weakly-ionized argon plasma, reveal a hexagonal crystal structure. The crystal is visible to the unaided eye. The particles are cooled by neutral gas to 310 K, and their charge is $>9,000 e$, corresponding to a Coulomb coupling parameter $\Gamma > 18,000$. For such a large Γ value, strongly-coupled plasma theory predicts that the particles should organize in a Coulomb solid, in agreement with our observations.

A low-power argon discharge at 2.05 ± 0.05 mBar was formed by applying a 13.56-MHz signal to the lower electrode of a parallel-plate reactor. The lower electrode is a disk 8 cm in diameter, while the upper is a ring, with inner and outer diameters of 3 cm and 10 cm, respectively. The electrode separation is 2 cm. The dc self-bias of the lower electrode was -14.5 ± 0.5 V, measured at the electrical feed-through. The rf power was 0.4 ± 0.3 W.

A few milligrams of particles were placed in a sieve above the hole in the upper electrode. This is agitated to release particles into the plasma. The particles are $7.0 \mu\text{m}$ diameter melamine/formaldehyde spheres, with a size distribution of $\pm 0.2 \mu\text{m}$.

The particles were seen as a thin disk-shaped cloud, levitated above the center of the lower electrode, near the sheath-plasma boundary. Particles were organized in approximately 18 planar layers (≈ 6 layers of two-particle agglomerates and ≈ 12 layers of single particles), parallel to the electrode. The diameter of the cloud was approximately 3 cm. Observations were made using a sheet of HeNe laser light to illuminate a plane. The sheet has a thickness of $110 \pm 23 \mu\text{m}$ and a breadth of 2 cm. It is adjustable to various heights above the lower electrode. Scattered light is viewed at 90° through a hole in the upper electrode. Individual particles were easily seen with the unaided eye and with a CCD video camera fitted with a macro lens. A 200-mm macro lens with extension tubes provided $\times 1$ magnification for the data shown below, while a long-distance microscope lens with a $\times 13$ magnification revealed that the particles in the plasma were either individual grains or agglomerates of two grains. The latter were found to populate the lower lattice planes of the cloud, presumably due to their greater mass to charge ratio.

An image of the particle cloud in a single plane reveals an organized structure with nearly uniform particle spacing. The image area is $7.7 \times 7.7 \text{ mm}^2$ and contains 724 particles. Assuming equal particle spacing in all directions, this corresponds to $n_p = (4.3 \pm 0.2) \times 10^4 \text{ cm}^{-3}$.

A numerical analysis of this image verifies it as a "plasma crystal." We identified the particle locations and to describe the Voronoi (Wigner-Seitz) cells). The cells are mainly six-sided, with nearly equal areas and uniform spacing. The mean separation Δ (lattice constant) between the particles is $250 \mu\text{m}$. The measurements were compared with a synthetic random particle distribution using the same properties - number and density - as in the experiment, This simulates a dusty plasma in its "gas phase".

This result showing the structure is from a single video frame. The dynamics of this structure are revealed by the video, which showed the individual particles oscillating gently about fixed equilibrium centers in what appeared to be a Brownian motion. Occasionally a grain diffused through the structure, visibly perturbing the position of the nearest particle.

In order to estimate the coupling parameter, Γ , we need to know the particles' kinetic energy (or temperature) and charge. The particles are cooled by the neutral gas. Based on frame-by-frame measurements of the mean particle velocity, we estimate the particle kinetic temperature to $T_p = 310 \text{ K}$, which is close to room temperature. Particles were also found to be at room temperature in the discharge of Boufendi et al. [9], who used laser-Doppler velocimetry to detect particles that were much smaller than ours, and reported a correspondingly smaller value of Γ .

We estimate the grain charge to lie between the extreme values $-9,000 \geq Q_p/e \geq -28,000$. The shielding length is estimated to lie in the range $60 \mu\text{m} \leq \lambda \leq 400 \mu\text{m}$, resulting in $4 \geq \kappa \geq 0.6$. The

Coulomb coupling parameter Γ is then estimated to be >18000 . Our observation of a crystalline structure is consistent with this large value of Γ .

NEED FOR MICROGRAVITY

Our experiment reveals that the dust cloud is crushed. It is much wider than it is high. This is thought to be due to a combination of gravity and the ion drag force. For spheres of the size we are using, it is inevitable that gravity will be a significant, and likely dominant, factor in this crushing.

To avoid the crushing, the *global electric force* that confines the grains in the plasma must be stronger than gravity. This is necessary so that particulates will be distributed uniformly throughout the vacuum vessel and don't pile up at the bottom due to gravity. Quantitatively, the potential energy difference across the vacuum vessel, Mg_0l , must be much smaller than the confining electric potential in the main field-free plasma region, $Q_p\phi$ which is $\approx Q_p k_B T_e / e$. Thus, the microgravity acceleration g_0 must satisfy

$$g_0 \ll Q_p k_B T_e / e M_p l$$

Now assuming a 20- μm radius plastic sphere, $T_e = 1 \text{ eV}$, and a floating potential of -10 V , this reduces to $g_0 \ll 0.667 \text{ cm s}^{-2} = 7 \times 10^{-3} \text{ g}$.

A second reason that gravity can disrupt crystal formation is that it competes with the *local electric field*. To achieve 3-D crystallization, the gravitational must be small compared to the local pressures on a grain. Electrostatic pressure is responsible for lattice formation, while gravitational pressure can inhibit it by crushing the lattice into a 2-D structure.

To summarize the anticipated microgravity requirements:

- The experiment requires $\ll 7 \times 10^{-3} \text{ g}$ of reduced gravity.
- Parabolic flights provide $\approx 10^{-2} \text{ g}$ of microgravity, which will be inadequate to assure a uniform density of spheres in the plasma.
- Spaceflight will provide $< 10^{-4} \text{ g}$, which is adequate.

CONCLUSIONS

Ground-based definition studies now in progress are revealing the soundness and feasibility of microgravity experiments for forming 3-D crystalline structures of charged microspheres levitated in a charge-neutral plasma. These experiments are expected to offer new opportunities for basic physics (Wigner lattice studies) and engineering (dust contamination during microelectronics processing).

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