

Effect of ion streaming on particle–particle interactions in a dusty plasma

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Dust particles in low-temperature, low-pressure plasmas form Coulomb crystals and display collective behavior under select conditions. The trajectories of ions can be perturbed as they pass by negatively charged dust particles and, in some cases, will converge beyond the particle. This process, called ion streaming, produces a positive potential in the wakefield of the particle that can be large enough to perturb interparticle dynamics. In this paper, we discuss results from a three-dimensional model for dust particle transport in plasma processing reactors with which we investigated the effects of ion streaming on particle–particle interactions. When including the wakefield potential produced by ion streaming, dust particles can form vertically correlated pairs when trapped in electrical potential wells. The ion-streaming force was found to be significant only over a select range of pressures and for given combinations of particle sizes and mass densities. The formation of vertically correlated pairs critically depends on the shape of the potential well. Wakefield forces can also affect the order of multilayer lattices by producing vertical correlations between particles in adjacent layers. © 2005 American Institute of Physics.
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I. INTRODUCTION

Dust particle transport in partially ionized plasmas has been investigated to provide insights to particle contamination of wafers during plasma processing of microelectronic devices and as a means to study nonideal plasmas.^{1–6} Particles in plasmas negatively charge to balance the flux of electrons and ions to their surfaces. As a consequence, the transport of these particles is governed by both mechanical (fluid drag, thermophoresis, gravity) and electrical (ion drag, electrostatic) forces. Particle–particle Coulomb interactions become important when the dust density is high enough that the interparticle spacing approaches the plasma shielding length, such as within a trapping site. For these conditions, particles interact through Debye shielded Coulomb forces. If the density of the particles reaches a critically high value, particles display collective behavior, a system typically referred to as a Coulomb solid or liquid. Observations of such behavior have been made in a variety of laboratory devices. For example, single layers of such interacting particles form hexagonal lattices.^{1–3} The interparticle spacing in these lattices may be a function of position if the magnitude of the compressive forces varies.³

When the particle count is large and the potential well is deep, more complex three-dimensional assemblies of dust particles have been observed with a range of order from face- and body-centered cubic lattices to more amorphous

arrangements.^{4,5} Weakly interacting vertical strings of particles have also been observed.^{7,8} These phenomena are not easily explained on the basis of the repulsive Debye potential and may be, in part, attributed to ion streaming. Ion streaming results from positive ions flowing past and focusing beyond negatively charged particles, generating wakefields of positive space charge. The typical conditions are for particles to be trapped in horizontal layers near the edge of the sheath. Positive ions, accelerated by the presheath and the sheath, perpendicularly intersect these layers as the ions proceed towards the boundary of the plasma. Positive space charge then forms in the wakefield below individual particles trapped at the sheath edge. The space charge can be sufficiently large that negatively charged particles at lower heights are attracted to the location of the positively charged below a particle trapped at a greater height, thereby producing vertical correlations between the particles.

Evidence of ion streaming has been seen in recent experiments. Samsonov *et al.*⁹ observed a nonreciprocal attractive interaction between two particles of different masses. The interaction is a sum of two forces; the repulsive electrostatic force, which dominates at short distances, and the attractive ion-streaming force, which dominates at larger distances. Hebner *et al.*¹⁰ measured the attractive and repulsive components of the interaction forces. They found that the structure of the ion-wakefield-induced attractive potential was significantly different from a screened-Coulomb repulsive potential and was responsible for the instabilities observed in multilayer assemblies.

Lampe *et al.*¹¹ developed an expression for the ion shadowing force by deriving the ion momentum intercepted by the upstream particle. In this formulation, the ion shadowing

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force scales linearly with the ion density and is an inverse square force as is the bare Coulomb force. They observed that the range of attractive force diminishes in regimes of relatively low plasma density, high gas pressure, and small particle sizes. Recently, Khrapak *et al.*¹² investigated the relative magnitudes of the shadowing forces associated with ions and neutrals in the isotropic bulk plasma and the plasma sheath region. They obtained analytical expressions for the resulting interaction potential and noted that neutral shadowing is of minor importance under most conditions of interest.

In this paper, we discuss results from a three-dimensional model for dust particle transport in plasma processing reactors in which the consequences of ion streaming are included. A dust particle transport model was integrated into a plasma equipment model to facilitate this study. When we include the wakefield potential produced by ion streaming, particles can form vertically correlated pairs as they settle in trapping locations near the sheath edge. The ion-streaming force was found to be significant only over a range of gas pressures for given combinations of particle sizes and mass densities. The model will be described in Sec. II, followed by a discussion of our parametric results in Sec. III. Concluding remarks are in Sec. IV.

II. DESCRIPTION OF THE MODEL

To address particle transport in low-temperature plasmas the Dust Transport Simulation (DTS) has been integrated as a module into the Hybrid Plasma Equipment Model (HPEM).¹³ The integrated Dust Transport Module (DTM) in the HPEM has been described in detail in Ref. 14 and so will only be briefly discussed here. The HPEM is a two- or three-dimensional simulation consisting of three main modules. The Electromagnetic Module (EMM) calculates electromagnetic fields and magnetostatic fields. These fields are used in the Electron Energy Transport Module (EETM) where electron-impact source functions and transport coefficients are derived. Results from the EETM are transferred to the Fluid-chemical Kinetics Module (FKM) to determine plasma source and sink terms. The FKM solves continuity, momentum, and energy equations for charged and neutral species and Poisson's equation for the electric potential. The outputs of the FKM are then fed back to the EMM and EETM modules for updated computations. The process iterates until a converged solution is obtained.

In the integrated HPEM-DTM, dust particles are introduced into the reactor after plasma properties (e.g., densities of charged and neutral species) computed by the HPEM have reached a steady state. Then, after each iteration through the modules of the HPEM, electric fields, neutral and charged particle fluxes and densities, and temperature gradients are exported to the DTM. These quantities are then used to compute forces on the dust particles and integrate their trajectories. At the end of a call to the DTM, dust particle densities and their charge states are returned to the HPEM. The locations of the dust particles are binned on the same numerical mesh as used in the HPEM and the densities are distributed on the mesh using finite-sized particle techniques. The dust

densities and their charges are then used in the solution of Poisson's equation and in computing electron and ion transport.

The DTM is a three-dimensional module in which the trajectories of dust particles are integrated based on mechanical and electrical forces. The forces included in the module are ion drag, fluid drag, electrostatic, thermophoretic, Coulomb, self-diffusive, gravity, and Brownian motion. The particle's surface electric potential is obtained by balancing the negative and positive charged particle currents to the particle. The forms of the mechanical and electrical forces used in the DTM are discussed in Ref. 14.

The Coulomb repulsive force between particles is based on the particle's shielded plasma potential, which, using the Debye-Hückel form, is

$$\Phi(r) = \Phi_0 \frac{a}{r} \exp\left[-\frac{(r-a)}{\lambda_L}\right], \quad (1)$$

where r is the radial distance from the center of the particle of radius a , Φ_0 is the surface potential of the particle, and λ_L is the linearized Debye length,

$$\frac{1}{\lambda_L} = \sqrt{\frac{e^2}{\epsilon_0} \left(\frac{N_e}{kT_e} + \frac{N_I}{2E_I} \right)}. \quad (2)$$

N_e and N_I are the electron and ion densities. T_e and E_I are the electron temperature and ion energy.

In addition to the interparticle Coulomb forces and conventional ion drag terms, we also included a shadowing or ion-streaming force between particles. Perturbation of ion trajectories by adjacent particles produces an electric potential in the wakefield of the upstream particle, leading to an attractive force to nearby negative particles. Lampe *et al.*¹¹ derived an expression for the shadowing force in an isotropic plasma,

$$F_s = -\frac{3}{8} \frac{a^2 Z^2 e^2}{\lambda_I^2 d^2}, \quad \lambda_I = \left(\frac{4\pi N_I e^2}{T_I} \right)^{-1/2}, \quad (3)$$

where Z is the particle charge, T_I is the ion temperature, and d is the interparticle separation.

In an anisotropic plasma, the velocity vectors of ions have a net directionality. For example, ions entering the presheath are accelerated to speeds characteristic of the electron temperature in a direction perpendicular to the electrode. Entering the sheath proper, ions may then be accelerated to energies of the order 10–100 eV. It is, in fact, this anisotropic ion velocity distribution, which produces the angularly dependent wakefield charge distribution that results in vertical correlations between particles. The derivation of a general expression for the ion-streaming force including anisotropy was beyond the scope of this work. To capture the properties of such an anisotropic force, we made the following adaptation and approximation to the expressions of Lampe *et al.*

We assume that the total ion-streaming force \vec{F}_{ST} is the sum of the force due to isotropic thermal fluxes of ions, \vec{F}_S , and a force due to a directed anisotropic ion flux, \vec{F}_D . The direction of \vec{F}_S is parallel to the interparticle chord whereas

the direction of \vec{F}_D is the chord between the particle and the center of the wakefield charge distribution. The magnitude of \vec{F}_D should scale linearly with the magnitude of the anisotropically distributed positive charge that the directed ion flux creates relative to the isotropic ion flux.

We assumed that the isotropic ion-streaming force could be represented by a ring of positive space-charge density n^+ around the dust particle. The magnitude of the positive space charge is

$$n^+ = \epsilon_0 \frac{\partial^2 \Phi}{\partial r^2}, \quad \text{where } \frac{\partial \Phi}{\partial r} = \frac{F_S}{Q}. \quad (4)$$

We then approximated that the positive space-charge density is equivalent to a net charge N^+ residing at a distance R from the particle,

$$N^+ = \int_a^{a+\lambda_D} n^+ 4\pi r^2 dr, \quad (5)$$

$$R = \frac{\int_a^{a+\lambda_D} r n^+ 4\pi r^2 dr}{\int_a^{a+\lambda_D} n^+ 4\pi r^2 dr} = \frac{\lambda_D}{\ln\left(\frac{a+\lambda_D}{a}\right)}. \quad (6)$$

The magnitude of the space charge due to the anisotropic directed ion flux is then

$$N_D^+ = \frac{\phi_D}{\phi_R} N^+, \quad (7)$$

where ϕ_D and ϕ_R are the magnitudes of the directed and random thermal ion fluxes. The location of the anisotropic charge resulting from an anisotropic ion streaming past a particle at position \vec{r}_i is

$$\vec{r}_{N^+} = \vec{r}_i + \frac{\vec{\phi}_D}{\phi_D} R, \quad (8)$$

where $\vec{\phi}_D$ is the anisotropic ion flux vector. The total ion-streaming force on particle j having charge Z_j is then

$$\vec{F}_{ij} = F_S \frac{r_i - r_j}{|r_i - r_j|} + \frac{\vec{r}_{N^+} - \vec{r}_j}{|\vec{r}_{N^+} - \vec{r}_j|} \frac{N_D^+ Z_j}{4\pi\epsilon_0 |\vec{r}_{N^+} - \vec{r}_j|^2}. \quad (9)$$

For computational convenience, interparticle forces were included only if the particles were within a specified distance of each other. For the conditions of interest, λ_L and the equilibrium interparticle separation are a few hundreds of microns to 0.1 cm. The Coulomb repulsion force decays exponentially and the attractive ion-streaming force decays in a quadratic manner with distance. Both forces are expected to be negligible at distances greater than a few λ_L . Choosing a maximum interaction distance of $20\lambda_L$ enables inclusion of the relevant interactions between particles while avoiding unnecessarily large computational times. A parametric study in which the maximum interaction distance was varied from $20\lambda_L$ to larger values confirmed that there was no discernible difference in the equilibrium particle separations when using the larger values. The only difference was expending more computer time.

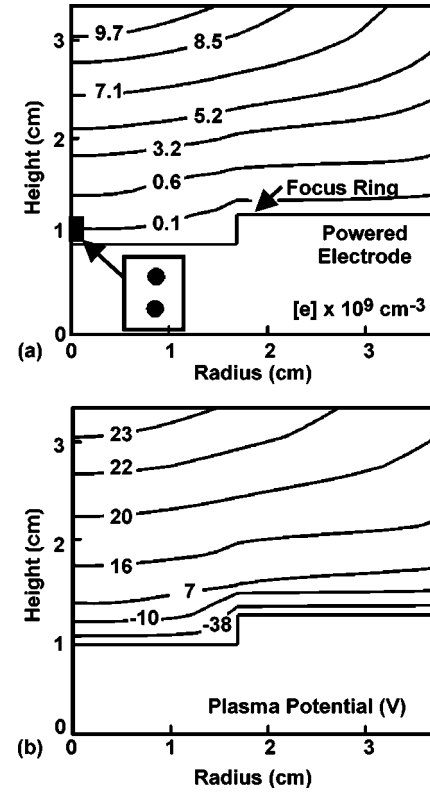


FIG. 1. Plasma properties in an Ar discharge (140 mTorr, 10 sccm, 10 W) in the vicinity of a well in the electrode. (a) Electron density (10^9 cm^{-3}) and (b) plasma potential (V). Dust particles settle into the minimum of the potential well shown by the shaded box.

III. PARTICLE INTERACTIONS WITH ION-WAKEFIELD FORCES

The reactor used in this work and the general plasma properties are similar to those discussed in Ref. 14. The reactor is a modified Gaseous Electronics Conference Reference Cell operated in a capacitively coupled mode.¹⁵ The lower electrode is powered at 13.56 MHz. A metal washer placed on the lower electrode acts as a focus ring to warp the electric potential into a well to confine the dust particles. Close-ups of the electron density and plasma potential in the vicinity of the potential well are shown in Fig. 1. The plasma was sustained in argon at 140 mTorr for a rf power of 10 W with a flow of 10 sccm (standard cubic centimeter per minute). The peak electron density and time-averaged plasma potential are 10^{10} cm^{-3} and 23 V, respectively. The dc bias on the substrate is -56 V . The ion density and plasma potential are conformal to the focus ring that forms a potential well in which the particles are trapped. The locations where the particles settle are generally on axis and are shown by the inset in Fig. 1.

To isolate the effects of ion streaming, we investigated the interaction between two particles having radii of 10 and 11 μm , and a mass density of 1.5 g/cm^3 . Dust particles are introduced into the reactor a few centimeters above the substrate and their trajectories were integrated until the particles settle into a quasistable geometric configuration. The side view of particle positions in the reactor is shown in Fig. 2. (The vertical line is the radius at which the potential of the trapping well is at its minimum height.) In the absence of the

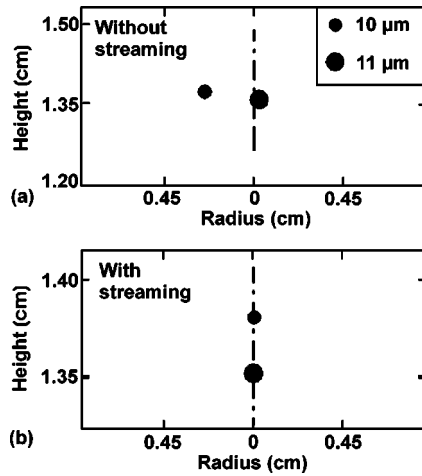


FIG. 2. Side view of particle positions in the Ar discharge (a) with and (b) without the ion-streaming forces for 10- and 11- μm particles.

attractive ion-streaming force, the interparticle force consists of only a shielded Coulomb repulsion. The particles come to rest at the edge of the sheath where ion drag and gravity are balanced by the opposing electrostatic force of the sheath. The larger particle sits slightly deeper into the sheath as the ion drag (and gravity) forces scale more acutely with radius than the opposing electrostatic force of the sheath. The minimum in the potential well is located between the particles which, in the absence of the repulsive Coulomb forces, is where both particles would sit. The repulsive Coulomb forces provide a radial offset that is more severe for the smaller particle.

When we include the ion-streaming force, the larger 11- μm particle, already sitting slightly below the 10- μm particle, is attracted to the region of the positive wakefield potential below the smaller particle. This results in a vertical correlation between the particles, as shown in Fig. 2. Both particles move to the location of the minimum of the electric potential, the larger particle under the smaller. The smaller particle is pushed upwards by the repulsive Coulomb forces from the larger particle until the larger particle sits in the minimum potential afforded by the positive wakefield charges.

Particle size has an important effect on the correlation between two particles interacting through wakefield forces. The sizes of the particles determine the vertical equilibrium position in the sheath and their displacement from the absolute minimum of the potential well. If the equilibrium positions of the particles are too far apart or their vertical displacements too small, the wakefield forces may be too weak to significantly perturb their positions. For example, we found less propensity for vertical correlations between a 9- and 11- μm pair than for a 10- and 11- μm pair. Smaller particles by virtue of their having smaller charges are less effective in deflecting streaming ions. This results in a smaller wakefield potential and, consequently, weaker interparticle attraction. The smaller particle also comes to rest higher in the sheath. Having too small a particle upstream therefore reduces the wakefield forces below a critical value required for vertically correlating a pair of particles. Also,

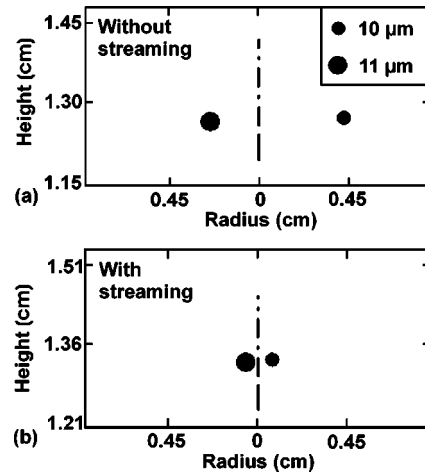


FIG. 3. Side view of particle positions in the Ar discharge (a) with and (b) without the ion-streaming forces for 10- and 11- μm particles when increasing their mass density to 2 g/cm³. This particle pair experiences a weaker attractive force.

the larger vertical separation between the 9- and 11- μm particle pair may prevent the 11- μm particle from experiencing the smaller wakefield potential below the 9- μm particle. A similar result was obtained by Hebner and Riley¹⁶ who observed that the attractive force between a 6.7- and 9.8- μm particle pair is almost twice that between a 6.7- and 11.9- μm particle pair.

To isolate the effect of vertical position in the reactor on ion streaming, we varied the mass density for the 10–11- μm particle pair. Particle positions are shown in Fig. 3. The electrostatic and ion drag forces depend only on radius, which determines their charge for a given set of plasma conditions, whereas denser particles settle lower in the sheath due to the increased gravitational force. The lower electron-to-ion density deeper in the sheath then results in smaller charges on the particles. Although the ion flux is nearly constant, the ion velocity increases deeper into the sheath thereby reducing the deflection around the upper particle and reducing the magnitude of the wakefield potential. The more dense particles do experience an attractive force with ion streaming which brings them closer together, but the force is not sufficient to vertically correlate them.

The pressure in the reactor can influence the ion-streaming effect in a variety of ways. For Ar at these operating conditions, the plasma density is higher at higher pressures, resulting in larger ion fluxes producing larger ion drag forces. On the other hand, the plasma is more collisional at higher pressures, and one would expect a weaker wakefield potential, as also noted by Lampe *et al.*¹¹ For example, at both 100 and 200 mTorr, ion-streaming forces were less significant than at 140 mTorr (Fig. 2). The heavier particle experiences a weaker attraction to the lighter particle, and the final particle positions are what one would expect in a system dominated by Coulomb repulsion alone. In a similar experiment, Hebner and Riley have observed that the peak attractive force between an 8.3- and 11.9- μm particle pair decreases from 230 to 160 fN on increasing the pressure from 60 to 100 mTorr.¹⁶ In any given system and shape of the potential well, there may be a narrow window of pres-

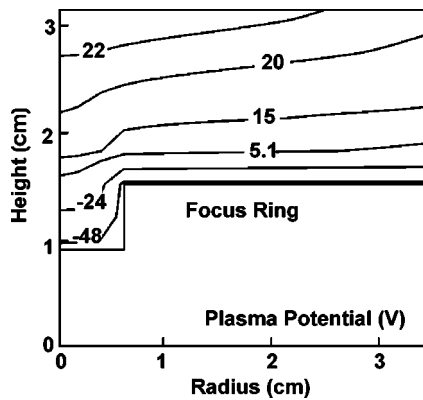


FIG. 4. Plasma potential (V) in an Ar discharge (140 mTorr, 10 sccm, 10 W) when using a deep potential well.

tures and powers for a given pair of particles, for which the equilibrium positions of the particles are close enough and the ion wakefield force large enough to vertically align the particles.

The formation of a vertically correlated pairs of particles also critically depends on the shape of the potential well. For example, we were able to obtain vertically correlated structures by using a physically deep potential well having a large aspect ratio even when excluding the ion-streaming force. In such a potential well, as shown in Fig. 4, the particles have a smaller radial extent they can access. As a result all particles will settle at or near the minimum of the potential well. Their mutual Coulomb repulsion then causes them to vertically stack with the larger particles on the bottom. Hence we observe correlated structures even when there is no attractive wakefield force between the two particles, as shown in Fig. 5, but a stronger vertical correlation when including the wakefield forces. The interparticle separation is 0.06 cm when we exclude the ion-streaming force and is 0.05 cm when we include it, indicative of the effect of an attractive wakefield potential. The particles with ion-streaming forces also physically sit deeper in the well. This may be a result of the pair of correlated particles acting like a single heavier particle.

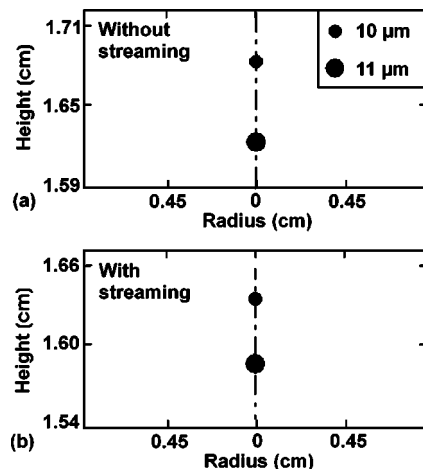


FIG. 5. Side view of particle positions in the Ar discharge with a deep potential well (a) with and (b) without the ion-streaming forces for 10- and 11- μm particles.

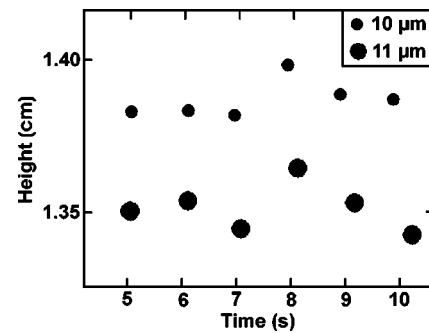


FIG. 6. Side view of particle positions at different times as they settle in a potential well defined by a focus ring in an Ar discharge (140 mTorr, 10 sccm, 10 W). Particles with different sizes (10 and 11 μm) form vertical pairs and exhibit correlated motion.

Recently, Hebner *et al.*¹⁰ measured the attractive and repulsive components of the interaction force from an analysis of trajectories of dust particles undergoing oscillation in a well-defined electrostatic potential. They used a shallow trench on the lower electrode, which formed an electrostatic trough in the plasma to confine the particles to one-dimensional lateral motion. Two particles released in the trough appeared to have correlated motion corresponding to an attractive ion-wakefield potential and a repulsive pairwise interaction.

In our computational approximation of the experiment of Hebner *et al.*, a metal washer placed on the lower electrode acts as a focus ring to warp the electric potential into a well to confine the dust particles. The motion of two particles as they settle in this potential well is shown in Fig. 6. On reaching the center of the well, the particle pair exhibits vertical correlated oscillations before they settle into a stable configuration. (The initial oscillations result from the seeded particles falling into the potential well from greater heights and overshooting their equilibrium positions.) The lower particle trapped in the ion wakefield of the upper particle follows the upper particle as it moves up in the potential well. The plasma conditions at the final position produce a larger charge for the upper particle and more repulsion and this leads to a larger interparticle separation.

Large assemblies of particles in plasmas may produce Coulomb fluids or crystals. Smaller numbers of particles typically form one layer crystals. Larger numbers of particles may form multilayer structures. Particles in a given layer experience a compressive electrostatic force from the potential well, as well as gravity, pushing them radially inwards. Coulomb forces between the particles keep them apart. Often there is a radially outward ion drag force which produces voids in the lattice.^{14,17} The balance of these forces results in particles settling in a disk-shaped configuration. Particles in adjacent layers (above and below) introduce an out-of-plane Coulomb repulsion and a wakefield attraction if ion streaming is taken into account. The interaction of particles in multiple layers was investigated with a Coulomb crystal of 100 particles, 50 each of sizes 10 and 11 μm . The side and top views of the particle positions when excluding the ion-streaming force are shown in Figs. 7(a) and 7(b). The particle positions (top view) in the individual layers are shown in

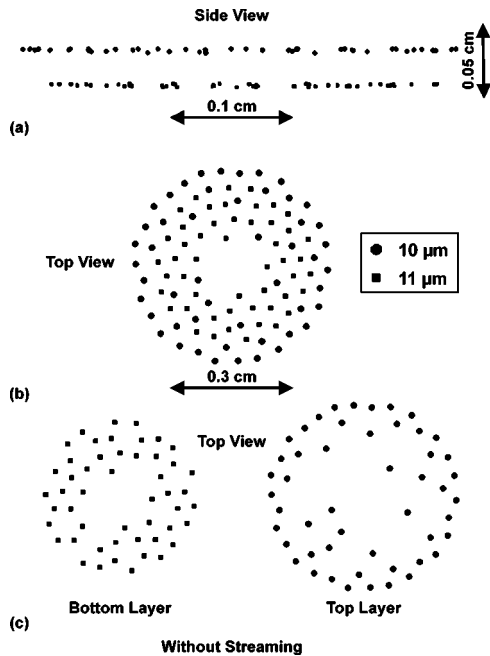


FIG. 7. Plasma crystal in an Ar discharge (140 mTorr, 10 sccm, 10 W) with fifty 10- and fifty 11- μm particles when excluding ion-streaming forces. (a) Side view, (b) top view, and (c) top view of the individual layers of particles.

Fig. 7(c). The corresponding particle positions when including the ion-streaming force are shown in Fig. 8.

The particles segregate into two layers with the larger particles constituting the lower layer. When viewed from the top, the lattice has an almost hexagonal structure in the absence of the wakefield forces. This structure results, however, from the superposition of two poorly structured lattices populated largely at outer radii by the smaller particles and inner radii by the larger particles. On the average the gaps in the upper layer are occupied by particles in the lower layer

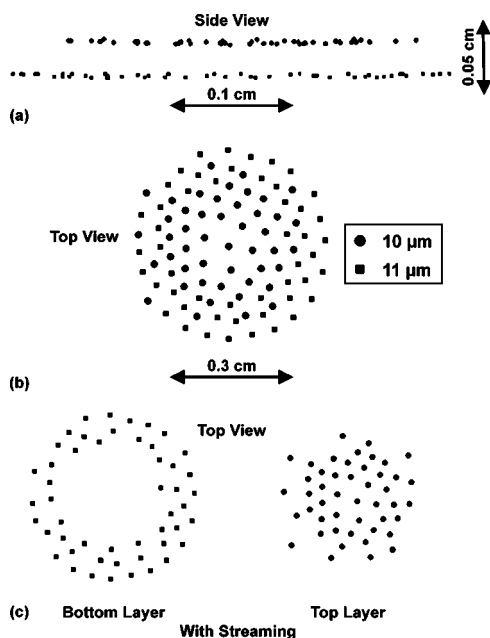


FIG. 8. Plasma crystal in an Ar discharge (140 mTorr, 10 sccm, 10 W) with fifty 10- and fifty 11- μm particles when including ion-streaming forces. (a) Side view, (b) top view, and (c) top view of the individual layers of particles.

(and vice versa). The vertical distance between the layers is small enough that the particles respond to the lateral Coulomb forces from particles in the other layer. The end result is a slightly disordered hexagonal lattice due to there being different sized particles and there being a vertical displacement.

When including the ion-streaming forces, the lattice, when viewed from above, appears less ordered. The larger particles now sit in the outer ring and the smaller particles assemble into a more disklike structure at smaller radii. The nearly equidistant hexagonal spacing obtained in the absence of ion streaming is now perturbed by the attractive wakefield forces between particles in the upper and lower lattices. The lateral spacings between 10- μm particles in the upper layer and adjacent 11- μm particles in the lower layer are smaller than spacings between 10- or 11- μm particles in the same layer. These vertical correlations result from the ion-streaming forces that produce a positive potential beneath the 10- μm particles in the top layer which then attract particles in the lower layer. Had there been only two particles, the ion-streaming force would have been strong enough to vertically correlate the particles (as shown in Fig. 2). In the confines of the lattice, the ion-streaming forces are only strong enough to displace the lower particles from what would have been their hexagonal equilibrium positions. The repulsive Coulomb forces of other particles in the lower layer prevent the particle from assuming a position directly under a particle in the upper layer.

IV. CONCLUDING REMARKS

A three-dimensional model has been developed to investigate particle transport and Coulomb crystal formation in plasma processing reactors. On including the forces due to ion streaming, particles can form vertically correlated pairs and show correlated motion as they settle in a potential well. Ion-streaming forces have a finite vertical extent and so their influence is ultimately determined by the equilibrium positions of particles in the absence of the forces. The streaming force was found to be significant only over a select range of pressures and for certain combinations of particle sizes and mass densities. In this regard, the formation of vertically correlated pairs critically depends on the shape of the potential well. In multilayered Coulomb crystals, the ion-streaming forces may produce disordering due to displacement of particles from their equilibrium hexagonal positions.

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