Response of an Isolated Particulate Subjected to Lateral Electric Fields in a DC Glow Discharge

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Abstract—Isolated spheres of borosilicate glass are suspended in a dc glow discharge in neon. The isolated sphere is laterally displaced by applying a transient voltage across a pair of electrodes mounted on the wall of the discharge tube. By monitoring the response of the particulate when subjected to a step change in the voltage and to sinusoidally varying voltages of different frequencies, the charge on the suspended sphere is inferred. Unlike most previous studies, this paper considers heavier (from 16 to 42 times heavier) particulates and heavily damped conditions. Measurements are reported for 20.3- and 4.9-μm-diameter borosilicate glass spheres. It is found that lateral excitation of an isolated particulate in a dc glow discharge cannot drive the motion to classical resonance as has been observed in previous work involving axial displacements in RF plasmas. The classical forced damped oscillator model used successfully in previous work to explain experimental observations is found to be inadequate for the conditions of this paper. Rather, the base excitation (BE) model is found to exhibit good agreement with the present experimental results at frequencies exceeding the frequency at which the displacement is a maximum. Sheath polarization, neglected in previous work, is included in this paper. It is found that drag due to collisions with neutrals is insufficient to account for the total drag on the particulate under heavily damped conditions. A new means of estimating the charge from the frequency response of the particulate motion using the BE model is described. Based on this method, charge numbers of $Z = 9.2 \times 10^3$ and $Z = 1.5 \times 10^4$ are inferred for the 20.3- and 4.9-μm spheres, respectively.

Index Terms—Plasma measurements, plasma sheaths, plasmas.

NOMENCLATURE

$A$ Amplitude of base motion.
$\Delta_P$ Particulate diameter.
$e$ Charge on an electron.
$E_{0a}$ Local axial electric field.
$E_a$ Ambipolar field.
$f$ Hertzian frequency.
$F_{\text{drag}}$ Drag force acting on particulate.
$F_r$ Restoring force acting on particulate.
$J_0$ Zeroth-order Bessel function.
$k$ Spring constant.
$k_b$ Boltzmann’s constant.
$k_p$ Spring constant representing restoring force due to polarization of particulate-adjacent sheath.
$m_n$ Mass of neutral atoms in plasma.

$I. \hspace{1pt} \text{INTRODUCTION}$

Solid particulates of any material can be suspended in a plasma by applying an electric field in the same direction as gravity. Such suspensions are possible because the particulates usually assume a negative charge in the plasma. Particulates that are suspended in sufficient proximity to each other form the lattice centers of solidlike structures having macroscopic dimensions ranging from millimeters to centimeters [1]–[3]. Typical particulate sizes range from micrometers to tens of micrometers, and distances between particulates are on the order of hundreds of micrometers. Various crystalline habits have been observed in RF discharges, while only amorphous and hexagonally close-packed structures have been observed so far in striated dc discharges. Before the interparticulate forces in a Coulomb solid can be understood, the behavior of a single isolated particulate must first be characterized. The focus of this paper is on the characteristics of a single isolated particulate in a dc glow discharge plasma.

Existing work on the behavior of a single suspended particulate may be classified into two categories. In the first category, one particulate within a solid or a cluster of two or more particulates is displaced and the resulting response is examined [3]–[7]. This paper falls into the second category in which an isolated single particulate is perturbed, and its response is studied [8], [9]. A majority of existing work has focused on particulates and Coulomb solids in low-pressure RF plasmas.
In contrast, only limited experimental results have been reported on the response of a single particulate displaced from a Coulomb solid suspended in a striated dc discharge [6]. In most cases, the particulates are displaced either by using radiation pressure from a laser beam or by superimposing a sinusoidally time-varying voltage on the applied axial RF electric field that is used to suspend the particulates.

Particulates suspended in RF plasmas have been shown to exhibit a damped harmonic response when subjected to a periodic excitation in the vertical direction [3]–[5], [7], [9]. The same response is also observed in lateral excitation in an RF discharge [8]. The periodic excitation is introduced in two ways. The first involves superimposing a low-frequency voltage on one of the electrodes of the RF discharge, resulting in a periodic vertical displacement of the trapped particulate or particulates from their equilibrium position [3], [4], [7]. A variant of this method involves inserting a copper wire in the vicinity of an isolated trapped particulate [8]. A low-frequency voltage is then imposed on the copper wire, causing the trapped particulate to displace in the horizontal direction. These methods rely on displacing the particulate by applying an external field, which must necessarily interact with the plasma as well as the particulate. The second means of inducing oscillatory particulate motion is to use radiation pressure from a chopped laser beam [5], [7], [9]. This method has the advantage that the external force is directly applied to the particulate, without the intervening effects of the surrounding plasma. Earlier work has used a classical second-order forced damped oscillator (FDO) dynamic model to demonstrate that regimes can be found where resonance occurs for lightly damped conditions [4], [5], [7]–[9].

In these earlier studies, only the retarding force due to momentum exchange via collisions between the particulate and neutral atoms of the host gas, and the restoring force due to the local electric field in the plasma were considered. Any forces arising from the presence of other nearby particulates or from the interaction with charges in the plasma were ignored. By using particulates of known mass, the charge on the particulate can be inferred from analysis of the frequency response of its maximum displacement (referred to as the resonance method in [4], [5], [7], and [8]). It is important to point out that among these earlier works, only [8] and [9] consider the behavior of a single isolated particulate.

There is one set of experimental measurements reported in the literature on the forced motion of a particulate at the edge of a Coulomb solid suspended in a dc discharge [6]. In these experiments, radiation pressure from a laser beam (same technique as in [5], [7], and [9]) is used to impulsively displace a single particulate at the edge of a Coulomb solid. The trajectory of the particulate is then subsequently observed, and its charge is inferred from a force balance applied at the instant when the velocity is a maximum. At this instant, the restoring force due to the local ambipolar electric field is balanced by the resistive drag force arising from collisions between the particulate and neutral atoms of the host gas. Any forces on the particulate due to the presence of the nearby Coulomb solid are ignored. This method is inherently different from the resonance method applied to study particulate motion in RF discharges. However, the accuracy in using this method to infer the particulate charge is susceptible to the same limitations as the resonance method because knowledge of local plasma conditions (e.g., average electron energy, ionization fraction, etc.) is needed [4], [5], [7], [8].

In this paper, measurements are reported on the response of a single isolated borosilicate glass sphere when subjected to a lateral (horizontal) force in a striated dc glow discharge in neon. This force is exerted by a lateral electric field externally applied by means of electrodes placed at the chamber walls. Either an abrupt step change in voltage, or a sinusoidally time-varying voltage is imposed on one of the electrodes, and the response of the particulate is measured. Unlike earlier work where lightly damped conditions were considered, this paper focuses on heavily damped conditions. A single-degree-of-freedom model of the particulate response is developed to interpret the experimental results. The model is then used to infer the charge on the particulate from the frequency response of the maximum particulate displacement. This paper considers particulates that are much larger and heavier than those considered in most previous work, as well as particulates with masses comparable to those in previous work.

This paper is organized as follows. The experimental apparatus and procedure are described in the next section, followed by a description of the experimental results in Section III, including measured responses of the particulate to step and sinusoidal excitations. Discussion of these results is given in Section IV. The base excitation (BE) model, used to interpret the results for sinusoidal excitation, is discussed in Section IV as well. Using this model and the experimental measurements presented in Section III, the charge on the particulates is inferred. A summary of this paper and its main conclusions are then given in Section V.

II. EXPERIMENTAL APPARATUS AND PROCEDURE

A schematic of the experimental apparatus is shown in Fig. 1. The apparatus consists of a 50.8 mm (2 in) diameter Pyrex glass tube, mounted vertically with its axis aligned with gravity. A very slow continuous downward flow (i.e., in the direction of gravity) of neon gas is used to maintain a steady-state pressure on the order of 25 Pa in the tube. The apparatus is equipped with two hollow cylindrical electrodes spaced 279.4 mm (11 in) apart along the axis of the tube. The electrode at the top is the anode, and the bottom electrode is the cathode. A discharge is struck inside the tube by applying a dc potential of 450–500 V across the axial electrodes. At the pressures used in the present experiments, the discharge is striated with stationary luminous zones separated by darker regions. Under these conditions, the discharge is near-extinction so the plasma is expected to be very weakly ionized. Particulates are introduced into the discharge from a dispenser located at the top of the tube, above the hollow anode. The dispenser is equipped with a 635 wire/in mesh sieve (Newark Wire Cloth Company) to allow only one or a few of the particulates to fall into the discharge with the slight turn of a handle. The particulates used in the experiments are monodisperse borosilicate glass spheres (Duke Scientific Corporation) with a density of \( \rho = 2500 \text{ kg/m}^3 \). Results are reported here for spheres of two different diameters: 20.3 ± 1.4 \( \mu \text{m} \).
In the experiments described here, a single isolated particulate is trapped in a striation. The discharge voltage, current, and pressure are selected such that the striation in which the particulate is trapped is leveled with the two lateral electrodes, as shown in Fig. 1. The particulate is thus situated between the 25.4 mm (1 in) diameter metal electrodes flush mounted on opposite sides of the wall of the tube. These lateral electrodes are located 139.7 mm (5.5 in) above the cathode, as shown in Fig. 1. The electrodes are used to displace the particulate from its equilibrium position by application of a lateral electric field.

This lateral electric field is imposed by applying a time-varying voltage on the left electrode (when viewed from the charge-coupled device (CCD) camera in Fig. 1) with respect to the right electrode. The electrical circuit that supplies this bias across the lateral electrodes is powered by 9-V batteries, thereby isolating it from Earth ground, the discharge, and the power supply that drives the discharge.

Two types of applied voltage transients are considered in this paper. In the first type, the voltage on the left electrode is varied as a step change to a constant value with respect to the electrode on the right. This step voltage transient has a duration of 16.4 s, with a rise time of about 50 ns and a decay time of about 500 ns. In the second type of transient, the voltage on the left electrode varies sinusoidally with a specific frequency. It is particularly important to note that the largest applied lateral electric field is on the order of 1 V/cm, which is much smaller than the magnitude of the axially applied field (>16 V/cm).

The particulate is illuminated by an unfocused low-power frequency-doubled YAG laser beam, and its motion is recorded on a CCD camera mounted at an angle 45° from the horizontal. The laser beam is directed perpendicular to the axis of the lateral electrodes. The laser is mounted such that it can be scanned vertically and laterally to ensure that there is only one particulate suspended. The capture rate of the CCD camera is approximately 17 Hz. Once the CCD images have been collected, the position of the particulate in each frame is determined using a two-step process. After digitizing the images, the particulate is pinpointed by setting all pixel values below a certain threshold to zero. Each particulate illuminates a field of several pixels. The centroid of this field of pixels is assigned the location of the particulate, and its coordinates are recorded. Time variation of the particulate motion relative to the applied lateral field is monitored using an LED mounted within the field of view of the camera. The LED is connected to the input circuit and is on when the sinusoidally time-varying voltage applied on the left electrode is positive, and off when it is negative. By this means, both the zero crossings of the applied voltage on the left electrode and the zero crossings of the equilibrium position by the particulate are simultaneously recorded. The difference between these zero crossings is defined as the phase shift between the particulate motion and the input voltage. The phase shift at each excitation frequency is taken to be the mean value over many of these successive differences between zero crossings. The root-mean-square (rms) value of the magnitude of the particulate motion is obtained from a fast Fourier transform (FFT) analysis of the particulate motion and is used to represent the magnitude of particulate displacement at a given excitation frequency.

For all the experimental data presented here, the magnitude of the average vertical displacement of the particulate is always significantly less than its lateral displacement.

### III. Experimental Results

Measurements of particulate motion when subjected to step and sinusoidally time-varying excitations are presented in this section. In the former case, a step change voltage transient is applied, and in the latter, a sinusoidally time-varying voltage is applied across the lateral electrodes. The sinusoidal excitation is applied at nine different frequencies: $f = 0.414, 0.619, 0.781, 0.930, 1.235, 1.384, 1.802, 2.809,$ and $4.255$ Hz. The measured quantities are the particulate displacement as a function of time and the phase shift of the particulate motion with respect to the sinusoidal excitation. These are described in the following sections.

#### A. Response of an Isolated Particulate to a Step Change in the Lateral Electric Field

The step transient consists of the sudden application of 6.7 V to the left electrode at a particular instant of time. Fig. 2(a) shows a time-lapse photograph of the typical displacement of a 20.3-μm-diameter particulate when subjected to this step transient. Fig. 2(b) shows the particulate displacement as a function of time obtained after digitizing each image. It can be seen from these figures that the negatively charged particulate initially displaces toward the positive (left) electrode as if it were subjected to an impulsive force (i.e., in the negative $x$-direction following the coordinate system established in Fig. 1). However, its motion is arrested at a particular position although the...
an impulsive force. Particulate displacement is shown here when a 20.3-μm borosilicate glass sphere is displaced by a step change in voltage to 6.7 V applied to the left electrode. This figure shows the typical quantitative data that is obtained after digitizing the image from each frame from the CCD camera. The data shown here is for a step change voltage transient to be approximately 170 ms.

The sharp displacement of the particulate to the left resembles a response to an impulsive force, although the lateral electric field is applied as a step change. The characteristic time for the particulate to attain its maximum displacement is measured from the step change voltage transient to be approximately 170 ms for the 20.3-μm particulate and 120 ms for the 4.9-μm particulate. The response time of the particulate is longer than that of the surrounding plasma because its inertia is orders of magnitude greater than that of the surrounding charged particles. The particulate slowly returns to its original equilibrium position because the potential at the center of the discharge tube is greater than the potential at the electrodes. This return process has a characteristic time on the order of 2 s since the restoring force is weak and continuously decreases as $x$ approaches zero at the center of the discharge tube, i.e., at the equilibrium position of the particulate before it was displaced.

**B. Response of a Particulate to a Sinusoidally Time-Varying Lateral Electric Field**

The typical response of an isolated borosilicate glass sphere when subjected to a sinusoidally time-varying lateral electric field is shown in the time-lapse photograph in Fig. 3. The figure shows the displacement of a 20.3-μm-diameter sphere when subjected to a sinusoidal voltage ($V(t)$) with a peak-to-peak amplitude of 11.56 V at a frequency of $f = 0.414$ Hz, applied on the left electrode. The figure shows clearly that the response of the particulate is sinusoidal, mimicking the imposed sinusoidal excitation. Once the image is digitized, the coordinates of the particulate $x(t)$ relative to its original equilibrium position.
are quantified. A sample of the typical data obtained from the digitized images is shown in Fig. 4, where the particulate displacement is plotted together with the variation of the applied voltage on the left electrode. Interestingly, the particulate attains its maximum displacement before the applied voltage reaches its maximum. This may be understood by recalling the results from Section III-A, which showed that the characteristic response time for the 20.3-μm particulate is 170 ms. Within this time, the local electric field in the vicinity of the particulate reverses direction, causing it to halt its motion toward the left electrode and turn back toward the center of the discharge. This behavior manifests itself in the positive phase shift between $x(t)$ and $V(t)$ as evident in Fig. 4. It is also shown in Fig. 4 that this positive phase shift establishes itself quickly, within the first half-cycle of applying the voltage on the left electrode, and persists thereafter. Data similar to that displayed in Fig. 4 are collected for different frequencies and amplitudes of the applied voltage. The maximum particulate displacement is obtained from the FFT of $x(t)$, and the phase shift is determined as described in Section II. Fig. 5 shows the FFT spectrum of the $x(t)$ displayed in Fig. 4. The frequency of the particulate response is clearly the same as the driving frequency. In the following section, these results are discussed and compared with a systems dynamics model, and contrasted with other experimental measurements reported in the literature.

Fig. 3. Time lapse photograph showing the response of a 20.3-μm borosilicate glass sphere when subjected to a sinusoidally varying voltage having a frequency of 0.414 Hz and an amplitude of 11.56 Vpp on the left electrode (see Fig. 1). The pressure is $P = 25.5$ Pa.

Fig. 4. Typical response of a 20.3-μm-diameter borosilicate glass sphere is shown for a sinusoidally time-varying voltage with frequency $f = 0.414$ Hz and 11.56 Vpp applied on the left electrode. The pressure is $P = 25.5$ Pa. Also shown in the figure is the temporal variation of the applied voltage imposed on the left electrode (solid curve). Particulate displacement is negative when it is toward the left electrode and positive when displaced toward the right electrode. Note that the particulate reaches its maximum displacement before the applied voltage reaches its maximum. This results in a positive phase shift between the two that is established within the first half-cycle from the instant the voltage is turned on and persists thereafter.

Fig. 5. FFT of the particulate displacement from Figs. 3 and 4 is shown here versus frequency. The sharp peak at 0.414 Hz indicates the frequency of the input sinusoidally time-varying voltage imposed on the left electrode.

IV. DISCUSSION

The response of an isolated particulate to sinusoidal excitation is examined in detail in this section. Measurements described in Section III-B are used to determine the frequency dependence of the particulate’s maximum displacement and its phase behavior relative to the applied voltage transient. A simple analytical model is developed to interpret the experimental measurements, and then used to infer the charge on the particulate.

Previous studies of either isolated particulates or particulates perturbed from a Coulomb solid in RF discharges have shown that their responses can be well represented as a simple FDO [3, 4, 7, 8] with a harmonic force applied directly to the mass. Fig. 6(a) shows a schematic of a systems dynamics model of this FDO where the particulate is represented by the mass $m_p$, the drag force is represented by the damper element with damping coefficient $\gamma$, and the restoring force is represented by
forces, this paper examines heavily damped conditions, unlike earlier work where resonance in the frequency response of the maximum particulate displacement was explored. Fourth, unlike some previous experiments in which the particulate is directly displaced using radiation pressure from a laser beam, the force due to the applied electric field in this paper acts on the particulate and the surrounding plasma or via the surrounding plasma. Finally, the phase shift component of the frequency response is explored as well as the magnitude of maximum particulate displacement.

A. Forces Acting on the Particulate

The behavior of a negatively charged particulate subjected to an electric field in a plasma has theoretically been considered in [10], where it is shown that while the charge cloud associated with the particulate shields it from the surrounding plasma, the particulate itself is not shielded from externally applied fields. It is therefore expected that interactions between the charged particulate and the surrounding plasma will result in a restoring force and damping force. In addition, the particulate is expected to encounter drag from the surrounding plasma both due to collisions with neutral atoms as well as retarding forces due to charged particle bombardment. The equation of motion of the particulate for the model shown in Fig. 6(a) may therefore be written as

$$m_p \ddot{x} = F_{\text{drag}} + F_t + F_{\text{ext}}$$

where an overdot denotes differentiation with respect to time, $F_{\text{drag}}$ is the retarding or drag force due to particulate collisions with the neutral atoms and due to particulate interactions with the charged particles in the surrounding plasma, $F_t$ is the effective restoring force due to the applied electric field and the surrounding plasma, and $F_{\text{ext}}$ is the externally applied force that induces particulate motion. The drag force arising from collisions between the particulate and neutral atoms of the gas is well known and may be written for the case where the particulate diameter $d_p$ is much smaller than the mean free path as [11]

$$F_{\text{drag}} = -\gamma_{\text{Epstein}} \dot{x} = -\frac{1}{3} \delta \pi d_p^2 m_n n_n \left( \frac{8 k_B T}{m_n \pi} \right)^{1/2} \dot{x}$$

where $T$ is the heavy particle temperature (i.e., temperature of ions and neutrals), $m_n$ is the mass of the neutral atoms in the plasma, $n_n$ is their number density, $k_B$ is Boltzmann’s constant, and $\delta$ is the Millikan coefficient, a constant with values between 1 and 1.444, depending on the nature of the collisions between the spherical particulate and the neutral atoms of the gas [11]. The coefficient of $\dot{x}$ is denoted as $\gamma_{\text{Epstein}}$ after [11]. However, the actual damping coefficient $\gamma$ of the particulate-plasma system is expected to be greater than $\gamma_{\text{Epstein}}$ owing to additional drag arising from the asymmetric distribution of ions in the vicinity of the particulate. The net effect, therefore, is to increase the effective damping coefficient

$$|F_{\text{drag}}| = | - \gamma \dot{x} | > | - \gamma_{\text{Epstein}} \dot{x} |.$$
According to [6], even in a striated dc discharge, the lateral potential distribution is governed by ambipolar diffusion, and the resulting restoring force $F_r = -eZ E_a$ can be calculated from the approximate ambipolar field, which is dependent on the gradient in the electron number density

$$E_a = -\frac{\varepsilon}{n_e e} \frac{dn_e}{dx}$$

where $n_e$ is the electron number density and $\varepsilon$ is the average electron energy (in joules) in the discharge. In addition, sheath polarization effects must be considered. When both the effects of the ambipolar field and sheath polarization are included, the resulting expression as given in [10] is

$$F_r = -eZ E_a + \frac{e^2 Z^2}{16\pi \varepsilon_0} \left(\frac{e^2}{\varepsilon_0 k_B T_e}\right)^{1/2} \frac{1}{\sqrt{n_e}} \frac{dn_e}{dx}$$

where $-eZ$ is the charge on the particulate, $\varepsilon_0$ is the permittivity of free space, and $T_e$ is the electron temperature which is taken to be a constant in this paper. The particulate charge is a dynamic quantity but is taken here to be steady (static) and constant. Equation (5) is written here in terms of the electron number density, whereas it is given in terms of the electron Debye length in [10]. For the cylindrical discharge geometry of the present experiments, $n_e$ may be approximated by the ambipolar diffusion solution [6], [12]

$$n_e = n_0 J_0 \left(\frac{2.4x}{R}\right)$$

where $J_0$ is the zeroth-order Bessel function, $R$ is the radius of the discharge tube, and $n_0$ is the electron density at the center of the discharge. For $x \ll R$, $J_0$ can be expanded in a Taylor series about $x = 0$, i.e., $J_0(\xi) \approx 1 - (1/4)\xi^2$. Using this approximation in (6) and substituting for $n_e$ in (5) yields [6]

$$E_a = \left(\frac{2.4}{R}\right)^2 \frac{\varepsilon x}{2e}.$$  

Substituting the linearized form of (6) and (7) into (5) yields

$$F_r = -Ze \left(\frac{2.4}{R}\right)^2 \varepsilon \frac{\varepsilon^2 Z^2}{32\pi \varepsilon_0} \left(\frac{e^2 n_0}{\varepsilon_0 k_B T_e}\right)^{1/2} \left(\frac{2.4}{R}\right)^2 x
= -k_a x - k_p x.$$  

The restoring force (8) has been linearized under the approximation $x \ll R$ and results in two distinct spring constants $k_a$ and $k_p$. The first term in (8) is the usual restoring force due to the ambipolar field experienced by the particulate, while the second term arises from the polarization of the charge cloud associated with the particulate [10]. Accordingly, $k_a x$ is referred to as the force due to the ambipolar field, and the second term $k_p x$ is called the polarization force. Equation (8) is appropriate for the positive column of a glow discharge. However, the present experiments, like those of [6], are in a striated glow discharge. The presence of such striations changes the radial distribution of electron density so that (6) is no longer strictly valid. However, since particulate displacements are small relative to the diameter of the discharge tube and confined to the immediate vicinity of the axis, the linearized form of (6) resulting in (7) and (8) is applicable in the present case. This is supported by experimental measurements and numerical calculations that show that the radial distribution of the potential in a striated discharge at a given axial location and instant of time is qualitatively similar to that in a positive column, particularly in the vicinity of the centerline about which the potential is symmetric [12], [13]. Since particulate displacements in the present experiments are confined to the vicinity of the centerline of the discharge, linearization of the radial electric field is justified and any distortions in the radial electric field may safely be neglected [12]. Note that $k_p$ scales as $Z^2$, and since $Z$ scales with the mass $m_p$, $k_p$ must become increasingly significant for heavier particulates.

Combining (1), (3), and (8), the governing equation for the FDO model is given by

$$m_p \ddot{x} + \gamma \dot{x} + k x = F_{ext}$$

where $k = k_a + k_p$. In spite of the incorporation of additional damping and stiffness in (3) and (8), it is found that the FDO model given by (9) cannot represent the measurements reported in this paper. This is primarily because the externally applied voltage across the electrodes changes the lateral potential distribution in the plasma. This, in turn, causes the charges in the plasma to displace, which then exert a force on the particulate in addition to the electric force directly felt by the particulate. Therefore, unlike the model depicted in Fig. 6(a), the model shown in Fig. 6(b) is more representative of the present experiments in terms of the application of the force.

B. BE Model

In the model shown in Fig. 6(b), the particulate is represented by the mass $m_p$, and the massless base represents the surrounding plasma. The net effect of the applied sinusoidally time-varying voltage on the left electrode is represented as a harmonic input displacement of the massless base, which in turn exerts a force on the particulate. The mass and base are connected via a spring and damper. Since the applied force is represented as arising from the displacement of the surrounding plasma, i.e., the base, this model is referred to as the BE model. If $x$ is the instantaneous displacement of the particulate as measured from $x = 0$ at the center of the discharge tube, and $\eta$ is the time-varying input displacement of the base (i.e., plasma surrounding the particulate), then the equation of motion in the BE model is given by

$$m_p \ddot{x} + \gamma \dot{x} + k x = \gamma \ddot{\eta} + k \eta$$

where $k = k_a + k_p$. This equation can easily be obtained from Fig. 6(b) by applying Newton’s law to the particulate and the massless base representing the surrounding plasma separately, and then setting the two forces equal to each other. Note that the classical FDO model of [3]–[5], [7], and [8] is recovered when the right-hand side of (10) is replaced by a single harmonic force $F_{ext}$. It is important to emphasize that (10) cannot be reduced to the governing equation for the FDO.
In (12), the maximum value of $X(\omega)$ occurs at the resonant frequency $\omega_0 = 2\pi f_0$. This frequency can be found by setting $dX(\omega)/d\omega = 0$, which yields a relationship between $f_0$ and the total stiffness and damping coefficients, $k$ and $\gamma$.

$$\gamma^2 = \frac{2k^3}{m_1\omega_0^5} - \frac{2k^2}{\omega_0^3}.$$  \hfill (14)

Note that $\omega_0 \neq \omega_n$, i.e., the damped and undamped resonant frequencies are unequal. It is also interesting to note the difference between (14) and the corresponding condition obtained by maximizing the amplitude in the FDO model: $\gamma^2 = 2m_p k - 2m_p^2 \omega_0^2$. If (14) is substituted into (12) and (13), a more useful form of the BE model emerges.

$$X(\omega) = A \sqrt{\frac{1 + \left(\frac{2\omega^2}{\omega_0^2}\right)}{\left[1 - \left(\frac{\omega}{\omega_n}\right)^2\right] + \frac{\gamma^2}{k^2}}} \left[\frac{-\frac{m_1\omega^3}{k^2 \omega_0}}{\left[1 - \left(\frac{\omega}{\omega_n}\right)^2\right] + \left(\frac{\gamma^2}{k^2}\right)}\right].$$  \hfill (15)

$$\phi(\omega) = \tan^{-1} \left[\frac{-\frac{m_1\omega^3}{k^2 \omega_0}}{\left[1 - \left(\frac{\omega}{\omega_n}\right)^2\right] + \left(\frac{\gamma^2}{k^2}\right)}\right].$$  \hfill (16)

where $k$ is obtained from (8)

$$k = k_a + k_p = Z \left(2 \frac{A}{R} \right) \varepsilon \frac{Z e^2}{2\pi \varepsilon_0} \left(\frac{\varepsilon^2 n_0}{\cos \theta k_B T_e}\right)^{1/2} \left(\frac{2A}{R}\right)^2.$$

The essence of the BE model is captured by (15)–(17). In the present experiments, the mass of the particulate $m_n$ is known. By measuring the frequency response $X(\omega)$, the resonant frequency $\omega_0$ can be determined since it is simply the frequency at which $X(\omega)$ is maximum. The amplitude $A$ in (15) is also determined from experiment and is chosen so that the maximum in $X(\omega)$ matches the experimentally recorded maximum. The plasma properties are estimated in the vicinity of the particulate. Since the discharge conditions are comparable to those of [6], the average electron energy is taken to be $\varepsilon = 3$ eV, and the ionization fraction is taken to be $\alpha = 10^{-8}$, so that the electron number density is $n_0 = \alpha n_e$. Note that (15)–(17) are parameterized by the single quantity $Z$, the charge number on the particulate. Thus, (15) and (17) are simultaneous equations for the two unknowns $k$ and $Z$. A unique value of $Z$ can then be determined by comparing (15) and (17) to the experimental data with (16) serving as an independent check. The model assumes that the value of $Z$ is constant, therefore the particulate charges estimated by this method represent average values.

Fig. 8(a) shows the maximum displacements $X(\omega)$ at different frequencies for the 20.3-μm sphere at 24.5 Pa, for a peak-to-peak bias of $V_0 = 10.13$ V applied on the left electrode. As shown in Fig. 8(a), the resonant frequency is $f_0 = 1.5$ Hz ($\omega_0 = 9.42$ rad/s). Also, shown in the figure is the trend predicted by (15) for $A = 0.405$ mm and $k = 2.104 \times 10^{-9}$ N/m. From (14), the inferred value of the damping coefficient is $\gamma = 3.391 \times 10^{-10}$ N·s/m. The trend predicted by the FDO
model using the same values of $k$ and $\gamma$ is also shown in the figure.

Fig. 8(a) shows several key points. First, both model prediction and measurements show that the particulate motion is heavily damped. Second, the BE model agrees with the experimental measurements quite well at the higher frequencies, while the agreement is poor at the lower frequencies. This is to be expected since the model does not account for the rearrangement of the potential distribution within the plasma when the external lateral electric field is applied. As was mentioned in Section III-A, the characteristic time for the plasma-particulate system to respond to an externally applied electric field is 170 ms for the 20.3-$\mu$m particulate. The corresponding frequency of a sinusoidal transient with comparable characteristic rise time (the first quarter of its period) is $f \sim (1/4(0.17)) = 1.47$ Hz, which is close to $f_0 = 1.5$ Hz at which the maximum displacement is observed. As shown in Fig. 8(a), the particulate-adjacent plasma is unable to respond to the changing applied electric fields at the higher frequencies (i.e., time scales shorter than 170 ms) so that the particulate moves as the applied electric field dictates. This is also evident in the behavior of the phase at frequencies exceeding 1.47 Hz [see Fig. 8(b)]. The departure of the predictions of the BE model from the experimental measurements at the lower frequencies may be understood as follows. At the lower frequencies of excitation that correspond to time scales longer than 170 ms, the particulate-adjacent plasma has sufficient time to rearrange its potential distribution in order to counter the externally applied electric field. Consequently, the maximum displacement of the particulate is much smaller than the value predicted by the BE model. It is worth noting that certain measurements of maximum displacement reported in [4] and [5] (e.g., data for 30 Pa in [4] and for 70 Pa in [5]) are overpredicted by the classical FDO model [Fig. 6(a)] at frequencies below the resonant frequency. Finally, it can be seen that the FDO model of [3]–[5], [7], and [8] fails to predict the experimental trends displayed in Fig. 8(a) for all values of the frequency $f$. In contrast, the BE model predicts the measurements shown in Fig. 8(a) well for $f > f_0$.

The phase angle $\phi$ between the particulate displacement $x(t)$ and the sinusoidally varying externally applied voltage is shown in Fig. 8(b) for the same conditions as Fig. 8(a). Also shown in this figure is $\phi$ calculated using both (16) and the FDO model. Consistent with the comparison in Fig. 8(a), it can be seen that the phase angle predicted by the BE model matches the measured values quite well for $f > 1.5(f_0)$ Hz. The positive phase shift at the lower frequencies may be better understood when viewed in conjunction with Fig. 4. When the positive bias is first applied to the left electrode, the particulate displaces toward it. However, as soon as the potential distribution within the plasma is able to rearrange itself so that the plasma potential at the center of the discharge is higher than the potentials at the electrodes, the particulate slows down and begins returning to its original equilibrium position while the applied voltage on the left electrode is still climbing toward its maximum. This is the source of the phase shift that establishes itself within the first cycle of the transient and which persists thereafter for subsequent cycles of the applied voltage. Note from Fig. 8(b) that agreement between (16) and the measurement is best for $f > f_0$. Moreover, both the measurements and predictions of the phase angle appear to asymptote at the higher frequencies. Equation (16) suggests that $\phi(\omega)$ should asymptotically approach $-\pi/2$ as $f \to \infty$. To the best of our knowledge, no previous work has reported on measurements of the phase angle of the particulate response relative to the excitation.

Fig. 9(a) and (b) compares measured and predicted $X(\omega)$ and $\phi(\omega)$ at different frequencies for an isolated 4.9-$\mu$m-diameter sphere at 26.5 Pa for a peak-to-peak bias of $V_0 = 11.41$ V applied to the left electrode. As shown in Fig. 9(a), the resonant frequency is $f_0 = 1.6$ Hz ($\omega_0 = 10.05$ rad/s). The predicted $X(\omega)$ and $\phi(\omega)$ from both (15) and (16) and the FDO model for $A = 0.415$ mm and $k = 3.222 \times 10^{-11}$ N/m are also shown in the figure. From (14), the inferred value of the damping coefficient is $\gamma = 4.688 \times 10^{-12}$ N·s/m. As in the case of the
The inferred charge for the 20.3-μm-diameter borosilicate glass sphere in neon at $P = 26.5$ Pa is $Z = 15 000 \pm 6750$. The uncertainty in $Z$ is determined as before, by using the root sum square method. The total uncertainty amounts to $\sim 45\%$ of $Z$. For this value of $Z$, the corresponding values of the system parameters are $k_p = 3.218 \times 10^{-11}$ N/m, $\gamma = 3.600 \times 10^{-14}$ N/m, and $k_p = 4.688 \times 10^{-12}$ N/s/m. It can be seen that the spring constant is determined almost entirely by the ambipolar restoring force, and sheath polarization effects are negligible for the smaller sphere. From (2), the contribution of neutral drag to the damping coefficient is $\gamma_{\text{drag}} = 3.138 \times 10^{-12}$ N·s/m, which accounts for about 67% of the total damping coefficient $\gamma$. The value of $\gamma$ for this case is 1.05.

C. Comparison With Previous Work

The results presented here clearly show that the heavier particulate has larger values of $Z$, $k$, and $\gamma$. The classical second-order FDO model is unable to predict the measurements reported here. In contrast, the BE model represents the measurements reported in this paper quite well for frequencies exceeding $f_0$. It is pertinent, therefore, to examine how well the BE model compares with previously reported measurements, such as those of [4]. Using the values of $k$, $\gamma$, and $\gamma_{\text{drag}}$, reported in [4] for a pressure of $30$ Pa, $X(\omega)/A$ is predicted by (15). Fig. 10 compares $X(\omega)$ predicted by the BE model [Fig. 6(b)] and the classical FDO model [Fig. 6(a)] with measurements reported in [4]. In both models, $A = 0.12$ mm. It can be seen that the BE model agrees quite well with both the classical FDO model and the experimental measurements of [4]. Similarly, when compared with the measurements reported in [8] and [9] for much smaller values of $\varepsilon$, the BE and FDO models are virtually indistinguishable, and the comparison is therefore not shown here. In other words, both models predict the experimentally measured resonance curves of [8] and [9] extremely well. It is interesting to note that the experimental value of $f_0$ obtained in this paper for the 20.3-μm-diameter borosilicate glass sphere is comparable to that reported in [8] for a 6.9-μm-diameter melamine formaldehyde sphere, whereas the values of $k$ reported in [8] are nearly two orders of magnitude smaller.
This is because of the disparity in the masses of the two particulates \( (m_p = 1.10 \times 10^{-11} \text{ kg} \) in this paper versus \( m_p = 2.60 \times 10^{-13} \text{ kg} \) in [8]).

Table I compares the experimental conditions considered in this paper with those of [4], [5], and [8]. Also, shown in the table are the inferred values of particulate charge from the respective experimental measurements as well as the values of \( \frac{\gamma}{m_p} \) (based on total drag coefficient) compared with the corresponding values of \( \gamma_{\text{Epstein}}/m_p \) (based on the drag coefficient due to collisions of the particulate with neutral atoms alone), the total spring constant \( k \) and the damping ratio \( \zeta \). This paper differs from previous work in several aspects. First, in contrast to most previous work, this paper considers the behavior of an isolated particulate that is laterally displaced, in a direction transverse to gravity. This implies that the restoring forces in our experiments are much smaller than in previous work (except [8]), so that the previously reported steep resonance curves cannot be obtained for the experimental conditions considered here. Second, much heavier particulates (up to 42 times heavier than those listed in Table I) are studied here. The heavier particulates (20.3-μm spheres) used in this paper carry a much higher charge than those in previous work. Third, this paper shows that the overall drag coefficient is higher for the heavier particulates and higher compared to the drag coefficient due only to collisions between the particulate and neutral atoms of the gas [11]. The observed higher drag force on the particulate arises from its dynamic interaction with the asymmetric distribution of charges in the surrounding plasma. Note that the presence of a resonance peak is determined by the value of the dimensionless damping ratio \( \zeta \). Therefore, it is not simply the particulate mass, spring constant or coefficient of damping that dictates whether a resonance peak appears prominently, but rather the combination of all three quantities.

The most comparable conditions between the present measurements and previous work are for the 4.9-μm borosilicate glass sphere \( (m_p = 1.54 \times 10^{-13} \text{ kg}) \) and the measurements of [8] for a 6.9-μm melamine formaldehyde sphere \( (m_p = 2.60 \times 10^{-13} \text{ kg}) \). Despite the different gases used (neon in this paper and krypton in [8]) and the different pressures \((\sim 26 \text{ Pa here versus } 4 \text{ Pa in [8]})\), the resonant frequencies are the same \((f_0 = 1.6 \text{ Hz})\). It can be seen from Table I that the values of \( k \) are reasonably close as well \((k = 3.222 \times 10^{-11} \text{ N/m versus } k = 4.83 \times 10^{-11} \text{ N/m in [8]})\). This is to be expected since [8] likewise uses lateral displacement of the particulate. However, the data of [8] exhibits a sharp resonance peak because their low operating pressure leads to a small value of \( \gamma \), which in turn yields a small value of \( \zeta \). Consequently, the conditions in [8] are lightly damped, in contrast to the heavily damped conditions in this paper. Unfortunately, a comparison of the values for the particulate charge \( Z \) reported in this paper and those of [8] is not possible since no values for \( Z \) are reported in [8].

Table I also shows that the results obtained here for the 4.9-μm borosilicate glass sphere are consistent with the results reported in [4] and [5] for the 9.48-μm diameter melamine formaldehyde spheres. Although the pressures used in this paper and that of [4] are quite close \((\sim 26 \text{ Pa versus } 30 \text{ Pa in [4]})\), the gas is different (neon here versus argon in [4]). Reference [4] reports a measured resonance frequency of \( f_0 = 15.4 \text{ Hz} \) versus the 1.6 Hz found in this paper. However, this is consistent with the fact that the corresponding values of \( k \) and \( m_p \) are drastically different \((k = 3.222 \times 10^{-11} \text{ N/m versus } 6.16 \times 10^{-9} \text{ N/m in [4]} \) and \( m_p = 1.54 \times 10^{-13} \text{ kg versus } 6.58 \times 10^{-13} \text{ kg in [4]} \)). The restoring forces in [4] and [5] are much larger than in this paper because of vertical versus lateral displacement of the particulates. Given the uncertainty of nearly 40% inherent in both measurements and the differences in \( k \) and \( m_p \) values, the different values of \( Z = 15000 \) in this paper and \( Z = 4200 \) reported in [4] are understandable.

Similarly, the results obtained here are consistent with those of [5]. The pressure and gas used in this paper are different from those used in [5] (neon at 26.5 Pa here versus helium at 42 and 70 Pa in [5]). Reference [5] reports measuring a resonance frequency of \( f_0 = 16.3 \text{ Hz} \) versus 1.6 Hz found in this paper. As in the case of [4], this is consistent with the fact that the values of \( k \) and \( m_p \) are quite different, as can be seen in Table I. Reference [5] reports a value of \( Z = 8320 \) at a pressure of 70 Pa in contrast with the value of \( Z = 15000 \) obtained here. This is understandable given the experimental uncertainty in the determination of \( Z \) and the differences in \( k \) and \( m_p \).

The charge on the particulate may also be determined using another method reported in [6], using the present measurements for step input excitation. In [6], the trajectory of a particulate perturbed from the edge of a Coulomb solid suspended in a striated dc glow discharge in neon is observed. The pressures in [6] are in the range of 66.7–200 Pa, while in this paper the pressure is 24.5–26.5 Pa. In [6], a melamine formaldehyde particulate is displaced out of a Coulomb solid by radiation pressure from a laser. The laser is abruptly turned off, and the particulate is allowed to return to its original equilibrium position. During its return, the particulate first accelerates due to the ambipolar restoring force and then decelerates due to drag. At the point of maximum velocity, it is assumed that the only forces acting on the particulate are the ambipolar restoring
force and drag due to collisions with neutrals. The charge $Z$ on the particulate can then be estimated by equating these two forces at the instant when the velocity of the particulate is a maximum

$$E_a e Z = \gamma_{\text{Epstein}} v_{nm}$$

where $v_{nm}$ is the maximum velocity of the particulate. Substituting for $\gamma_{\text{Epstein}}$ and $E_a$ in (18) using (2) and (7) yields the following expression for the charge on the particulate:

$$e Z = \frac{8 \pi}{3} \left( \frac{R}{2.4} \right)^2 \frac{2 \alpha^2 v_{th} m_n n_n v_m}{\varepsilon |x_m|}$$

where $|x_m|$ is the location at which the particulate velocity $v_m$ is maximum. For a voltage of 6.7 V suddenly imposed on the left electrode, images of the particulate trajectory have been recorded and digitized. Measurements reveal a maximum velocity of $3 \times 10^{-3}$ m/s at the location $x_m = -4.1 \times 10^{-4}$ m for the 20.3-$\mu$m sphere. Substituting these values into (19) yields $Z = 2.79 \times 10^5 \pm 1.63 \times 10^5$ for $\varepsilon = 3$ eV, a value comparable to $Z = 9.2 \times 10^5 \pm 4.14 \times 10^5$ obtained from the measurements for harmonic excitation. Given the uncertainties associated with each method, the agreement between the resulting values of $Z$ is quite good. There is a greater uncertainty in application of the method outlined in [6] (about 60%) for the present experiments mainly because of the larger uncertainty associated with the measurement of the maximum particulate velocity. The uncertainty in the position of the particulate is $\pm 1$ pixel ($\pm 43 \mu$m) in the CCD camera used to capture the images and the error in temporal measurement is $\pm 1$ frame at the 17 Hz capture rate of the camera, so that the total uncertainty in $v_{nm}$ is approximately 36%–46%. Combined with the uncertainty in the average electron energy $\varepsilon$, the total uncertainty in applying this method is greater than the resonance method of [3]–[5], [7], and [8], and the method described earlier in this section for the harmonic excitation case. The charge estimates reported in [6] range from $Z = 3000$ for a melamine formaldehyde particulate 1.87 $\mu$m in diameter ($m_p = 5.17 \times 10^{-15}$ kg), to $Z = 1.1 \times 10^5$ for a melamine formaldehyde particulate 13.57 $\mu$m in diameter ($m_p = 1.98 \times 10^{-12}$ kg). Therefore, although the dc discharges used in [6] operate at pressures higher than those used here (approximately 25 Pa for this paper versus 66.7 to 267 Pa for [6]), the charge estimates made using this method are consistent for both particulate sizes considered here.

### V. Summary and Conclusion

Isolated particulates suspended in a dc glow discharge have been horizontally displaced (i.e., transverse to gravity) by applying a transient voltage across two electrodes placed laterally across from each other on the walls of the discharge tube. The two voltage transients imposed on the left electrode are a step change in the voltage and a sinusoidally time-varying voltage. This differs from most previous works that have either considered an isolated particulate in RF discharges or a single particulate displaced from the edge or from within a larger Coulomb solid in RF and dc discharges. Particulates with mass comparable to previous work, as well as those with masses much greater than previously considered, have been studied in this paper. The conditions reported here correspond to heavily damped cases, in contrast with the lightly damped situations studied in previous work. With the aid of the BE model, the charge on the suspended particulate and associated quantities such as the total spring constant $k$ and total damping coefficient $\gamma$ have been inferred from measurements. In this paper, values of $Z = 9.20 \times 10^5 \pm 4.14 \times 10^5$ and $Z = 15,000 \pm 6750$ are obtained for the 20.3- and 4.9-$\mu$m-diameter borosilicate glass spheres, respectively. Sheath polarization, ignored in previous work, is shown to be significant for heavier particulates. Comparisons between the present measurements, previous measurements, the classical FDO model and the BE model reveal that:

1) the present measurements agree well with other existing measurements for an isolated particulate [8] and are consistent with other existing measurements made on single particulates that are constituents of a larger Coulomb solid [4], [5];

2) lateral excitation of an isolated particulate in a dc glow discharge cannot drive its motion to exhibit a sharp resonance peak as in previously studied RF plasmas because the restoring forces are smaller, leading to higher values of the damping ratio $\zeta$;

3) the classical FDO model of the particulate is adequate for describing previous experimental results involving lighter particulates and lightly damped conditions ($\zeta \leq 0.62$ in previous work).
4) the classical FDO model is found to be inadequate in describing the present experimental results involving both light and heavy particulates for heavily damped ($\zeta > 1$) conditions;
5) the BE model provides a good representation of particulate motion compared to the FDO model for both lightly and heavily damped cases, but only provided that $f > f_0$ for the latter case, where $f_0$ is the resonant frequency;
6) neither the BE model nor the classical FDO model appear to be capable of representing all experimental conditions because neither considers the initial response of the particulate-adjacent sheath or surrounding plasma when an external force is applied;
7) the present experimental measurements for heavily damped conditions show the importance of sheath polarization and additional drag arising from dynamic interaction between the particulate and surrounding plasma in determining particulate motion.

This paper clearly shows the presence of additional effects such as sheath polarization and dynamic interaction between the particulate and surrounding plasma, which are operative when heavier particulates are suspended in plasmas and subjected to heavily damped conditions. These are likely to be important for determining the structure, properties, and dynamics of Coulomb solids in plasmas as well. Dynamic models are found to represent some aspects of particulate motion in plasmas, but are far from complete. More complete models that include effects important for heavier particulates and heavily damped situations are needed before every aspect of the experimental measurements is explained and predicted. These models must necessarily consider the response of the particulate-adjacent sheath and surrounding plasma when an external force is applied to induce motion.

REFERENCES


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