Pulse excited oscillation: A new high-precision excitation method for the charge-to-mass ratio determination of microparticles in plasma and comparison to stepwise excitation and the phase-resolved resonance method

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Pulse excited oscillation: A new high-precision excitation method for the charge-to-mass ratio determination of microparticles in plasma and comparison to stepwise excitation and the phase-resolved resonance method

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ABSTRACT
The charge-to-mass ratio of microparticles confined in the sheath of an RF plasma is the key parameter for the understanding and theoretical description of dusty plasmas. Standard techniques, like the resonance method (RM) and the phase-resolved resonance method (PRRM) based upon the harmonic oscillator model of the microparticle, are used to determine the charge-to-mass ratio. However, if high precision is required, these methods become relatively slow. In this work, we present two transient response-based methods, the step excited oscillation method, adapted and modified from Meijaard et al. [Phys. Plasmas 28, 083502 (2021)], and the new pulse excited oscillation method (PEOM). A careful comparison to the PRRM and others is presented. The PEOM offers a significant increase in speed while maintaining a precision comparable to that of the PRRM.

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I. INTRODUCTION
The systems commonly used to study dusty plasmas are microparticles confined in the sheath of an RF plasma. A) Aside from the utilization of plasma crystals as a model system for strongly coupled systems, individual particles increasingly find application as minimally perturbative probes, for example, as diagnostics for the sheath potential shape, impulse flux in ion beams, or as thermal probes. Other methods like the rotating electrode method (REM) employ microparticles to access particle properties such as the charge as well as plasma parameters like the Debye length. To understand the behavior of the particle and its interaction with the surrounding plasma, knowledge of the particle properties is essential. One such property is the particle's charge-to-mass ratio $q/m$. Shortly after the successful generation of plasma crystals in the laboratory, the first resonance method for $q/m$ determination was presented. Since then, multiple methods to determine $q/m$ have been developed that rely on the modeling of the trapped particle as a harmonic oscillator. Instead of an electrostatic excitation as in the RM, the particle can also be excited by applying radiation pressure with a laser beam. An improvement over the resonance method, the phase-resolved resonance method (PRRM) allows for high-precision measurements of the eigenfrequency $\omega_0$ and neutral drag coefficient $\gamma$ of the particle and was successfully used to show the degeneration of plastic microparticles inside of the plasma sheath. This method, which can be called the “working horse” of high precision determination of $q/m$, has so far been successfully applied to the investigation of several research questions, such as the ion wake, charging of non-spherical particles, the mass- and density change of polymer particles in the plasma, and for the determination of $q/m$ in binary mixtures of microparticles. Unfortunately, the high precision of the PRRM is associated with a large time requirement, which is detrimental for time-critical processes like plasma-inherent etching of polymer particles. Recently, it was shown that $q/m$ can in principle also be obtained with a stepwise modulation of the RF power. This causes an attenuated oscillation.
motion of the particle, which can then be analyzed. Related methods have been used to examine particle pairs and their interaction potential\(^{27,28}\) or the shape of the electric potential in the sheath.\(^{29,30}\) In all these approaches, the excitation consists of a singular, practically instantaneous event, which causes a transient state of the particle as an oscillator to occur. The trajectory during this transient response is then evaluated according to the individual method. These methods are therefore called transient response methods. In contrast, the data of the RM and PRRM stems from a multitude of individual steady states arising from single-frequency sinusoidal excitation. The resulting resonance curve is a compound of all this information. Transient response approaches have the potential to be much faster than the PRRM and RM, because there is no need to vary the driving frequency of the driver, which is time-consuming because the resolution depends on the chosen frequency steps and the decay of transients. Instead, the step function (or a signal with similarly abrupt changes) used as an excitation signal contains theoretically all possible frequencies in its spectrum. The transient response methods can therefore also be called multi-frequency excitation methods, in contrast to the single-frequency excitation methods or steady-state response methods RM and PRRM.

In this work, we present a precise and fast method based on a pulsed excitation of single dust particles and critically compare it to the PRRM and stepwise excitation methods. Section II will cover the theoretical background, the concept of the different methods, and the necessary model functions for the evaluation of each method’s data. After describing the experimental setup and the measurement and analysis procedures in Sec. III, the findings will be presented and thoroughly discussed in Sec. IV, before a final conclusion is drawn in Sec. V.

II. MEASUREMENT CONCEPT AND CONSIDERATIONS

A. Forces on the microparticle

A dust microparticle in the sheath of an RF plasma attains a large negative charge \(q_{\text{st}}\) of several \(10^{2}\)–\(10^{6}\) elementary charges.\(^{31,32}\) As there is no cloud of other dust particles, there is first no interaction with other particles, and second the plasma suffers no electron depletion due to the Havnes effect.\(^{29,30}\)

The particle is subjected to multiple forces, which scale differently with the particle radius \(a\). Of these, the gravitational \((\propto a^2)\), electrostatic \((\propto a^2)\), and neutral drag forces \((\propto a^3)\) are dominant. Other forces are the thermophoretic force \((\propto a^3)\), which can be neglected because there is no measurable temperature gradient in the experiment, and the ion drag force \((\propto a^2)\), which is estimated using the models by Hutchinson and Khrapak as detailed in the textbook by Melzer.\(^{33}\) Using typical values for the sheath \((k_B T_e = 3 \text{ eV}, k_B T_i = 30 \text{ meV}, a = 3.7 \mu \text{m}, \text{ and } n_i = 1.2 \times 10^{19} \text{ m}^{-3})\), the ion drag is calculated to be at least one order of magnitude weaker than the gravitational and electrical force and therefore neglected in this work.

In mechanical equilibrium, the particle levitates at a height \(z_0\) determined by the balance between the gravitational force \(F_g = m \cdot g\) and the electrostatic force \(F_E = q_{\text{st}} \cdot E(z)\). In motion, the particle’s movement is attenuated by the neutral drag force \(F_{\text{drag}} = -\gamma \cdot z\). For spherical particles, this is described by the Epstein friction\(^{34}\) with

\[
\gamma = \frac{4}{3} \frac{p}{\pi \rho_{\text{ne}} \nu_{\text{th}, n}}
\]

Here, \(\rho_{\text{ne}}\) is the mass density of the particle material and \(\nu_{\text{th}, n}\) is the thermal velocity of the ambient neutral gas. \(\gamma\) is a geometric coefficient, which value was shown to be 1.44 for microparticles in typical laboratory circumstances.\(^{17}\) Finally, the dust grain can be accelerated by an arbitrary external driving force \(F_{\text{ext}}(t)\). This system may be treated as a driven harmonic oscillator for small vertical displacements from the equilibrium position \(z_0\) because the potential is still parabolic, while nonlinear effects arise at higher amplitudes.\(^{32}\) This leads to the well-known equation of motion for the driven harmonic oscillator

\[
\dddot{z} + 2\gamma \dot{z} + \frac{q_{\text{st}}^2}{m \rho_{\text{ne}}} \dot{z} = \frac{1}{m} F_{\text{ext}}(t).
\]

Assuming that the height dependence of the particle charge is much smaller than that of the electrical field in the sheath, the eigenfrequency \(\omega_0\) of this system can be written as

\[
\omega_0 = \sqrt{\frac{q_{\text{st}}^2}{m \rho_{\text{ne}}}}
\]

\(\rho_{\text{ne}}\) is the absolute charge density at the position of the particle in the sheath. (The index \(q\) is chosen to avoid confusion with the mass density \(\rho_{\text{me}}\).) The RM and PRRM utilize this by evaluating the system’s (in the case of PRRM complex) resonance curve \(A(\omega)\) resulting from excitation with a sinusoidal frequency sweep to obtain the parameters \(\omega_0\) (eigenfrequency) and \(\gamma\) (see also Fig. 3). Another characteristic frequency worth mentioning is the resonance frequency sweep \(\omega_{\text{res}} = (\omega_0^2 - 2\gamma^2)^{1/2}\). It marks the maximum of the amplitude’s absolute value \(|A(\omega)|\) in the resonance curves of the RM and PRRM. On the other hand, the stepwise excitation method (called SEOM for step-excited oscillation method below), as well as the method presented in this work, the pulse-excited excitation method (PEOM), utilizes the response of the particle to a step- or (rectangle-) pulse-shaped excitation signal directly in the time domain. This response is a damped oscillation (see Secs. II B and II C) with a characteristic frequency \(\omega_d = (\omega_0^2 - \gamma^2)^{1/2}\), the damped eigenfrequency. It is important to differentiate between \(\omega_d\) and \(\omega_0\), as they are slightly different.

B. Excitation mechanisms

There are multiple ways to generate an electrical excitation (driving force) to this system, of which those two will be focused on that are found to be most practical in the laboratory situation. Both rely on distorting the sheath by a small extent and with it the equilibrium position \(z_0\). The first way, labeled as “modulated,” is by amplitude-modulating the RF power that generates the plasma with an additional low-frequency signal. This changes the spatial extent of the plasma and thus the equilibrium position of the levitating microparticle. One can reasonably assume that the signal modulation and the change in the plasma occur quasi-instantly relative to the timescale of the particle motion, which has a damped eigenfrequency of around 10 Hz. Thus, the resulting driving force is directly proportional to the modulation signal. The second way, labeled “bias” below, is by adding the excitation signal as a bias voltage to the driven electrode. This adds the need to incorporate a filter network (shown in Fig. 1) into the experimental setup in order to protect the hardware generating the low-frequency excitation signal against the RF voltage. The effect of this filter network
ard the Ohmic resistance, which depends on the discharge parameters. The impedance of the capacitors can be neglected, as the impedance of the capacitors $Z_C(\omega) = 1/(i\omega C)$ dominates for low $\omega$. There are multiple ways to determine the effect of the filter on the incoming signal. For periodic signals, this is usually done by determining the system’s frequency-dependent transfer function $H(\omega)$, which can be easily written by looking at the filter network as a voltage divider as follows

$$H(\omega) = \frac{U_e}{U_{LF}} = \left(1 + \frac{2C_2}{C_1} + \frac{\omega_0}{i\omega}\right)^{-1},$$

where $\omega_0 = 1/(R_p C_1)$ is the system threshold frequency, and $R_p$ is the Ohmic plasma resistance, which depends on the discharge parameters such as RF power and pressure. While this approach is well suited for methods like the PRRM\textsuperscript{1} that employ periodic signals, it is difficult to apply to the step- and pulse signals used in SEOM and PEOM. In this work, the system’s Green’s function is a much easier way to determine $U_e$ from $U_{LF}$. By analyzing the circuit diagram with Kirchhoff’s circuit laws, one can first derive a first-order ordinary differential equation for the in- and output voltages $U_{LF}$ and $U_e$,

$$\frac{d}{dt} + \nu U_e = c U_{LF},$$

with $c = (1 + 2C_2/C_1)^{-1}$ and $\nu = c/(R_p C_1)$. $\nu$ is related to $\omega_0$ as $\nu = c \cdot \omega_0$. Because the plasma resistance $R_p$ contained in $\nu$ is the only unknown variable of the filter network, $\nu$ is introduced as a fit parameter into the fit models for the bias methods. Because the capacities $C_1$ and $C_2$ are known, $R_p$ can then be calculated.

In Eq. (5), only the derivative $U_{LF} = \frac{dU_{LF}}{dt}$ of the input voltage $U_{LF}$ appears, so the filter acts as an analog differentiator for low-frequency signals. Using the Fourier transform of Eq. (5) and the residue theorem, one can easily write Green’s function $G_i(t)$ for the differential operator in Eq. (5) as

$$G_i(t-t') = \theta(t-t') \cdot e^{-\nu(t-t')}.$$  

This allows us to calculate the response to any input signal $U_{LF}(t)$ as

$$U_e(t) = \int_{-\infty}^{\infty} G_i(t-t') \cdot c U_{LF}(t') \, dt'.$$

In particular, we can use this to model the driving force $F_{ext}(t)$ and obtain the particle’s response to $F_{ext}(t)$ using Green’s function $G_p$ of the particle as a harmonic oscillator (which is routinely derived in many mathematical textbooks\textsuperscript{33})

$$G_p(t-t') = \theta(t-t') \cdot e^{-\nu(t-t')} \cdot \sin\left(\frac{\omega_0(t-t')}{\omega_0}\right).$$

This response function can then be used as a fit function $\xi(t)$ to extract the parameters $\omega_0$ and $\gamma$, among others.

### C. Model functions

In addition to the PRRM, which will act as a reference method, there are three closely related excitation methods examined in this work. Their characteristic excitation signal shapes, particle responses, and the corresponding behavior of the plasma RF voltage are shown in Fig. 2.

#### 1. Modulated SEOM (moduSEOM)

Meijaard et al.\textsuperscript{26} introduced the SEOM with a power modulation in the form of a Heaviside step function $\theta(t)$. For a single step excitation, the cycle-averaged RF power $P_{RF}$ is modeled as

$$P_{RF}(t) = P_1 \pm \theta(t) \cdot \Delta P,$$

where $P_1$ is the initial power, and $\Delta P$ is a power step-up- or downward occurring at $t = 0$, resulting in a power of $P_e = P_1 \pm \Delta P$. In the experiment, this is realized by applying a slow square signal ($T = 2s$) and evaluating the response to each slope. One has to consider that the
oscillator parameters obtained are those corresponding to the RF power after the slope, as the plasma parameters and particle charging take place on timescales < 0.5 ms, which is about three orders of magnitude faster than the particle oscillation (T ≈ 100 ms). We can then assume that the change in \( z_0 \) can be reasonably treated as instantaneous, and the external driving force on the particle can be written as

\[
F_{\text{ext}}(t) = B \cdot \theta(t),
\]

with \( B \) being the resulting amplitude of the force step. The particle response can then be modeled with the following fit function:

\[
z(t) = z_0 + D \cdot e^{-\gamma t} \cdot \sin \left( \omega_d t \right),
\]

where \( D \) is the initial amplitude containing the direction of the slope and the resulting initial motion in its slope.

2. Bias-SEOM (biasSEOM)

As discussed above, the filter necessary in experimental setups with bias excitation distorts the excitation voltage signal \( U_i \). For a Heaviside-step \( U_i(t) = U_b \cdot \theta(t) \), the resulting driving force is easily calculated [because \( U_i(t) \propto \theta(t) \)] using Eq. (7) and is written as

\[
F_{\text{ext}}(t) = B \cdot e^{-\gamma t} \cdot \theta(t).
\]

Using the oscillator’s Green’s function [Eq. (8)], one can obtain the particle’s response

\[
z(t) = z_0 + D \cdot \left[ e^{-\gamma t} \sin \left( \omega_d t - \phi \right) + b \cdot e^{-\gamma t} \right],
\]

with the quantities \( b = \left[ 1 + ( \nu - \gamma )^2 / \omega_d^2 \right]^{-1/2} \) and \( \phi = \arctan \left( \omega_d / (\nu - \gamma) \right) \). This is a damped oscillation superimposed with an exponential decaying offset due to the filter network. A sufficient oscillation amplitude can be reached with a far smaller perturbation of the plasma, as the input excitation amplitude \( U_b \) is only 1 V compared to the peak-to-peak RF voltage of about 60 V and the self-bias of roughly –50 V. The slope direction still influences the resulting parameters but not nearly as strongly as in the modulated case.

3. Bias-PEOM (biasPEOM)

The next improvement consists of using a rectangle pulse with a duration \( t_p \) instead of a single step as an excitation as follows:

\[
U_i(t) = U_b \cdot \left( \theta(t) - \theta(t - t_p) \right).
\]

Exploiting linearity, we can construct our driving force

\[
F_{\text{ext}}(t) = B \cdot \left( e^{-\gamma t} \cdot \theta(t) - e^{-\gamma (t-t_p)} \cdot \theta(t-t_p) \right)
\]

and particle response from the biasPEOM case

\[
z(t) = z_0 + D \cdot \left[ e^{-\gamma t} \sin \left( \omega_d t - \phi \right) + b \cdot e^{-\gamma t} \right. \\
\left. - e^{-\gamma (t-t_p)} \sin \left( \omega_d (t-t_p) - \phi \right) - b \cdot e^{-\gamma (t-t_p)} \right].
\]

From the resulting fit parameters, the eigenfrequency can be calculated as

\[
\omega_d = \sqrt{\omega_d^2 + \gamma^2}.
\]

While the precision for \( \omega_d \) is mathematically dependent on both \( \omega_d \) and \( \gamma \), it can be easily verified using the basic error propagation that it is primarily given by the precision of \( \omega_d \) for \( \gamma \ll \omega_d \). Under the experimental circumstances in this work, \( \gamma < 0.09 \omega_d \) was found, and it is thus assumed that the uncertainty in \( \gamma \) has only a small influence on the uncertainty of the calculated \( \omega_d \).

The pulse duration \( t_p \) can be chosen deliberately. In this work, we conducted the biasPEOM with pulse durations of 10, 50, and 100 ms. A simple consideration regarding this is that the amplitude should reach its maximum when the acceleration takes place, while the velocity is at its maximum and in the same direction as the acceleration. The second slope should thus occur after roughly half a period \( T_d \) of the particle’s oscillation, so that the external pulse acceleration is approximately in phase with the particle’s oscillation. This leads to a criterion for the pulse duration \( t_{p, \text{opt}} \) with the maximum resulting amplitude

\[
t_{p, \text{opt}} \approx \frac{T_d}{2} = \frac{1}{2 \omega_d} = \frac{\pi}{\omega_d}.
\]

The particles used in this work had damped oscillation frequencies close to 10 Hz, so \( t_p = 50 \text{ ms} \) was chosen as an optimal pulse duration. The shorter and longer pulse durations were chosen for comparison to assess the validity of the criterion Eq. (18).

III. EXPERIMENTAL PROCEDURE

A. Setup

The measurements are conducted in the StickingCube plasma chamber (shown in Fig. 3), a capacitively coupled RF discharge operated with argon at a pressure of 5.5–10 Pa. A flow controller is used to adjust the pressure in the vacuum vessel. The plasma setup consists of two circular electrodes with a diameter of 30 mm and a distance of...
34 mm, and the RF-driven lower electrode features a cylindrical cavity with a diameter of 10 mm and a depth of 2 mm. This cavity geometry produces an electric field component in the radial direction in the sheath, which confines the negatively charged microparticles horizontally above the center of the electrode. The upper electrode is grounded, while the lower electrode is driven by an RF generator outputting 1.5 W. The RF power can be modulated with an analog input connected to a computer interface. Additionally, a low-frequency bias voltage signal can be applied to the lower electrode via the aforementioned filter network. The total RF signal between the electrodes is monitored with an oscilloscope. Between the bias and modulated configuration, the self-bias voltage \( U_{sb} \) and the amplitude \( U_{RF} \) of the RF voltage do not change. However, while the power is raised to 1.9 W in the modulated case, the self-bias drops from about –50 to –55 V, and the RF voltage amplitude rises from circa 60 V peak-to-peak to almost 70 V peak-to-peak. During the bias excitation, no difference in the RF voltage amplitude rises from circa 60 V peak-to-peak to almost 70 V peak-to-peak. 

A computer interface allows for analog and digital in- and outputs and is controlled via National Instruments LabVIEW for automated data acquisition. It outputs all the required excitation and timing signals and is used to monitor the pressure, RF amplitude, and RF self-bias voltage. The excitation signals for PRRM, moduSEOM, biasSEOM, and biasPEOM are generated by a LabVIEW program and coupled into the discharge as either a modulation of the RF power or an additional bias voltage signal at the lower electrode after passing the filter network. A high-speed camera with a macro lens is triggered by a digital output signal. The camera has a resolution of 1440 \( \times \) 1080 pixel (16 megapixel) on a 4.97 \( \times \) 3.73 mm\(^2\) CMOS sensor, with a pixel size of 3.45 \( \mu \)m. For the PRRM, the trigger occurs four times per period of the excitation signal, while the other time series for the other methods are recorded at 200 frames per second. The particle is illuminated by a green laser (\( \lambda = 532 \) nm) with 250 mW that is expanded to circa 9 mm diameter using a beam expander optic, in order to illuminate the lower electrode, with a distance of 1 pixel corresponding to 0.38 \( \mu \)m. A computer interface allows for analog and digital in- and outputs and is controlled via National Instruments LabVIEW for automated data acquisition. It outputs all the required excitation and timing signals and is used to monitor the pressure, RF amplitude, and RF self-bias voltage. The excitation signals for PRRM, moduSEOM, biasSEOM, and biasPEOM are generated by a LabVIEW program and coupled into the discharge as either a modulation of the RF power or an additional bias voltage signal at the lower electrode after passing the filter network. A high-speed camera with a macro lens is triggered by a digital output signal. The camera has a resolution of 1440 \( \times \) 1080 pixel (16 megapixel) on a 4.97 \( \times \) 3.73 mm\(^2\) CMOS sensor, with a pixel size of 3.45 \( \mu \)m. For the PRRM, the trigger occurs four times per period of the excitation signal, while the other time series for the other methods are recorded at 200 frames per second. The particle is illuminated by a green laser (\( \lambda = 532 \) nm) with 250 mW that is expanded to circa 9 mm diameter using a beam expander optic, in order to illuminate the whole area in which the particle oscillates. A mirror configuration allows for adjustment of the illuminated area. The LabVIEW program automatically detects the particle’s position on the camera image and saves it together with the corresponding timestamp. Using an image of a scale between the electrodes before or after the measurement, the pixel position of the particles is calibrated to the absolute height above the lower electrode, with a distance of 1 pixel corresponding to 4.74 \( \mu \)m.

B. Evaluation

Each of the 13 single particles was examined individually at 5.5, 7.5, and 10 Pa using all the methods (moduSEOM, biasSEOM, and biasPEOM with \( f_p = 10, 50, \) and 100 ms) consecutively before switching to the next pressure. For each particle and pressure, two PRRM curves were acquired, as well as at least 20 repetitions of each other method. The resulting data for the PRRM were evaluated according to the standard procedure, and the corresponding results are shown in Fig. 4. For the other methods, the corresponding fit function (see Sec. II C) was fitted to the data using a nonlinear least squares algorithm.

The position data were then translated from pixel to millimeters, using a calibration with a scale as mentioned in Sec. III. Exemplary curves for every method are shown in Fig. 5. The eigenfrequency \( \omega_c \) could then be computed from the damping \( \gamma \) and the resonance frequency \( \omega_m \) using Eq. (17). Thus, these results are directly comparable to those of the PRRM.

IV. RESULTS

A. Statistical comparison

The results for \( \omega_c \) and \( \gamma \) are statistically analyzed and compared to the PRRM results. The ratio between each method’s results and the corresponding PRRM result is calculated and shown in Fig. 6. This normalization to the results of the PRRM is needed to eliminate the influence of the particle size distribution, as the particle radius \( a \) affects the particle charge and damping (see Sec. II). This size distribution stems from the small uncertainty with which the microparticles are produced by the manufacturer; see Sec. III for the exact numbers. The same information as in Fig. 6 is presented in Table I to show the exact numbers. In the following, it is important to differentiate the terms accuracy and precision: accuracy is the ability to obtain the true value of the measured quantity, which we assume the PRRM value to be, while precision describes the magnitude of the uncertainty arising while measuring the quantity.

The (relative) systematic deviation \( R_{sys} \) presented in Table I is (for a given method and pressure) given as

\[
R_{sys} = \frac{\bar{\gamma} - \bar{\gamma}_{PRRM}}{\bar{\gamma}_{PRRM}},
\]

where \( \bar{\gamma} \) is the mean value a given method yields at a given pressure, \( R_{sys} \) is used as a measure for the accuracy of the method. On the other hand, the relative statistical uncertainty \( R_{stat} \) measures the precision of a method and is given by
\[ \sigma_j = \frac{\sigma_y}{\gamma_{PRRM}}. \] 

(20)

\( \sigma_j \) is the standard deviation of all values the given method yields at a given pressure. Both these values are first determined for each given combination of pressure, method, and individual particle and then averaged over all individual particles. The corresponding values of \( R_{\text{sys}} \) and \( R_{\text{stat}} \) for \( \gamma \) and \( c \) are calculated in the same way. Generally, the uncertainty for \( c \) is about one order of magnitude smaller than that of \( \gamma \). All methods but the moduSEOM (which overestimates \( c \) by circa 4%) are capable of reproducing the PRRM result for \( \gamma \) very precisely (with deviations from the PRRM reference of at most 0.5%). The quality of the results seems to also be not dependent on the pressure. As a first finding, all bias methods deliver extremely accurate results for \( \gamma \) with the 50 ms-biasPEOM offering the best precision, with uncertainties of about 0.2%. The overall picture is similar for \( c \); the moduSEOM overestimates \( c \) significantly. However, the biasSEOM and the 100 ms-biasPEOM underestimate \( c \) by ca. 7%. The 10- and 50 ms-biasPEOM are accurate with a deviation from the PRRM values of \( \leq 2\% \). Of those two, the 50 ms-variant is decidedly more precise. This is likely due to the worse signal-to-noise ratio in the data from the 10 ms-biasPEOM because of the lower oscillation amplitude.

To further assess the plausibility of these results, Monte Carlo simulations using synthetic data\(^{36}\) for the 50 ms-biasPEOM were conducted, and predicted uncertainties on the order of 0.1% for \( \gamma \) and 1% for \( c \), agreeing well with the experimental results. These theoretical uncertainties are extremely close to those of the PRRM also acquired via synthetic data.\(^{18}\)

Interestingly, the moduSEOM results for \( c \) seem to be dependent on the pressure, from which the rest of the methods are largely independent. There are two possible explanations: first, the moduSEOM presumably produces more noise in the data by the strong perturbation of the plasma (especially in the relatively small plasma reactor) and has a slightly smaller oscillation amplitude than the bias methods. This unfavorable signal-to-noise ratio makes the accurate determination of gamma difficult. Another reason lies in the concept of excitation via modulation itself: Meijaard et al.\(^{26}\) stated that the stepwise method (corresponding to the moduSEOM in this work) is sensitive to the delayed charging effect (DCE) proposed by Pustylnik et al.\(^{37}\) The DCE basically adds another term to the Epstein friction coefficient in Eq. (2),
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| TABLE I. Results of the statistical analysis, equivalent to the data in Fig. 6. For each method and pressure, the relative systematic deviation $R_{sys}$ and statistical uncertainty $R_{stat}$ are presented. $R_{sys}$ corresponds to the vertical position of the data points in Fig. 6, while $R_{stat}$ corresponds to the size of the error bars.

<table>
<thead>
<tr>
<th>$\omega_0$</th>
<th>5.5 Pa</th>
<th>7.5 Pa</th>
<th>10 Pa</th>
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<tr>
<td>Method</td>
<td>$R_{sys}$</td>
<td>$R_{stat}$</td>
<td>$R_{sys}$</td>
</tr>
<tr>
<td>PRRM</td>
<td>... ±0.16</td>
<td>... ±0.09</td>
<td>... ±0.10</td>
</tr>
<tr>
<td>mSEOM</td>
<td>+3.7 ±0.54</td>
<td>+4.0 ±0.75</td>
<td>+3.7 ±0.90</td>
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<tr>
<td>bSEOM</td>
<td>−0.19 ±0.24</td>
<td>+0.13 ±0.32</td>
<td>+0.12 ±0.54</td>
</tr>
<tr>
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<td>−0.18 ±0.48</td>
<td>−0.03 ±0.53</td>
</tr>
<tr>
<td>bPEOM 50 ms</td>
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<td>−0.36 ±0.21</td>
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<tr>
<td>bPEOM 100 ms</td>
<td>+0.35 ±0.28</td>
<td>+0.11 ±0.23</td>
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<td>$\gamma$</td>
<td>5.5 Pa</td>
<td>7.5 Pa</td>
<td>10 Pa</td>
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<td>$R_{stat}$</td>
<td>$R_{sys}$</td>
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<td>−</td>
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<tr>
<td>bPEOM 10 ms</td>
<td>−2.5 ±6.3</td>
<td>−1.8 ±6.3</td>
<td>−9.1 ±6.0</td>
</tr>
<tr>
<td>bPEOM 50 ms</td>
<td>−2.9 ±2.1</td>
<td>−2.2 ±2.7</td>
<td>−3.8 ±2.6</td>
</tr>
<tr>
<td>bPEOM 100 ms</td>
<td>−7.0 ±5.6</td>
<td>−7.3 ±3.7</td>
<td>−8.4 ±3.1</td>
</tr>
</tbody>
</table>

\[
\gamma = \gamma_{\text{eps}} + \gamma_{\text{DCE}}. \tag{21}
\]

This term $\gamma_{\text{DCE}}$ is, among others, dependent on the pressure. Should this effect be of non-negligible magnitude, it could explain why $\gamma$ is measured as so much higher by the modusSEOM than the other methods, as well as its apparent pressure dependence.

B. Optimal pulse duration

Regarding the criterion in Eq. (18), it can be said that the 50 ms-biasPEOM, satisfying the criterion, indeed delivers the highest overall oscillation amplitude using the same excitation amplitude and thus the best signal-to-noise ratio. The criterion for the pulse duration is therefore regarded as reasonable. Looking at Fig. 5(c), one can see that a very complicated trajectory arises when the criterion is grossly violated and the excitation is in antiphase with the particle oscillation. Such trajectories can be harder to evaluate or yield less precise results. It should be noted that to fulfill the criterion, an estimate of $\omega_0$ is needed. This can be, for example, based on a very coarse manual frequency sweep with a sinusoidal excitation and a high exposure time of the camera, similar to the classical resonance method (RM) or simply from experience.

C. PEOM supporting PRRM, access to plasma resistance $R_p$

Another advantage of the bias methods is that they also provide access to the plasma resistance $R_p$ via the fit parameter $\nu_i$ which is usually difficult to obtain accurately with the PRRM due to the weak dependency of the resonance curve on it. This, combined with the time resolution (1–2 s per measurement) of these methods, allows for another way of monitoring the plasma state, e.g., to ensure the reproducibility of experiments. It would also be possible to use the PEOM as a quick diagnostic before conducting a PRRM measurement: The results for $\gamma$ and $\omega_0$ would help define the frequency range of the PRRM measurement, and the result for $\nu$ could be used to improve the PRRM fit; the PRRM fit contains the fit parameter $\omega_0$ as the critical frequency of the filter in the frequency domain. This parameter is, due to the aforementioned weak effect on the shape of the resonance curve, difficult to determine via a PRRM fit. $\omega_0$ is related to $\nu$ as

\[
\omega_0 = \nu \cdot \left(1 + \frac{2C}{C_1}\right) . \tag{22}
\]

Thus, from the PEOM result for $\nu$, one can get a very good estimate for the parameter $\omega_0$ in the PRRM fit. It could be incorporated as a fixed value for $\omega_0$ as a well-defined start value, or to set bounds for the fit parameter.

D. Determination of the charge-to-mass ratio $q_d/m$

Equation (3) allows us to determine the charge-to-mass ratio $q_d/m$ of a dust particle from its eigenfrequency $\omega_0$ when the absolute charge density $\rho_q$ in the sheath is known. Using typical values (ion density $n_i = 1.2 \times 10^{14} \text{m}^{-3}$, electron duty factor $n_e/n_i = 0.3$), the charge density is $\rho_q = 8.4 \times 10^{13} \text{e/m}^3 \approx 1.35 \times 10^{-5} \text{C/m}^3$. Table II shows the resulting charge-to-mass ratio and the charge of an exemplary particle ($2a = 7.38 \mu$m) for different discharge pressures.

E. Final assessment

Taking into account all these findings, the biasPEOM is deemed the optimal method among those examined in this work. This is attributed to the following qualities:

- The excitation with an additional bias voltage is only a very small distortion of the plasma (especially in comparison to the excitation via an RF power modulation).
- The rectangle-pulse shape of the excitation signal is also beneficial, as choosing a pulse duration according to the criterion in Eq. (18) further aids in obtaining low-noise data for the particle trajectories.
- The incorporation of the filter characteristics into the fit model yields an extremely good agreement between data and fit model and also allows to gain access to the plasma resistance $R_p$.
- As a result of the last three arguments, the biasPEOM is comparable to the PRRM in terms of precision and reproduces its results with a

| TABLE II. Variation of the change-to-mass ratio and the charge of an exemplary ($2a = 7.38 \mu$m) particle with the discharge pressure. The values for $\omega_0$ from the PEOM measurements were used. The mass density of SiO$_2$, $\rho_m = 1850 \text{kg/m}^3$ is needed to compute the particle charge.

<table>
<thead>
<tr>
<th>Pressure $p$/Pa</th>
<th>Eigenfrequency $\omega_0$/rad $\cdot$ s$^{-1}$</th>
<th>Charge-to-mass ratio $\frac{q_d}{m}$/10$^{-22}$ C kg$^{-1}$</th>
<th>Charge $q_d/e$</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.50</td>
<td>68.48</td>
<td>4.94</td>
<td>7560</td>
</tr>
<tr>
<td>7.50</td>
<td>75.46</td>
<td>6.00</td>
<td>9180</td>
</tr>
<tr>
<td>10.0</td>
<td>85.17</td>
<td>7.65</td>
<td>11700</td>
</tr>
</tbody>
</table>
high accuracy. It also outperforms the biasSEOM with its slightly simpler step-excitation in regard to the uncertainties in $\gamma$.

V. CONCLUSION

A high-precision method, the biasPEOM, for the determination of $\alpha_d$ and $\gamma$ of a trapped microparticle was developed, thoroughly tested, and compared with existing, similar methods like the moduSEOM (stepwise excitation). The rectangle-pulse signal shape and the excitation with a bias voltage in conjunction with the incorporation of the filter network in the model allow for a very precise analysis of the data. The high time resolution potentially allows for better monitoring of time-dependent processes, for example, the etching of polymer microparticles in argon or argon–oxygen plasmas. Additionally, the ability to determine the plasma resistance $R_p$ can be used to monitor the plasma state. It also qualifies as a supplement to PRRM measurements. Furthermore, the biasPEOM operates with precision comparable to the PRRM and reproduces its results reliably with good accuracy, also justifying the use as a stand-alone diagnostic for $\alpha_d$ (via $\alpha_d$) and $\gamma$ that is much faster than the PRRM.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Armin Mengel: conceptualization (equal); data curation (lead); formal analysis (lead); investigation (lead); methodology (equal); project administration (equal); software (lead); validation (lead); visualization (lead); writing—original draft (lead); and writing—review and editing (equal).

Maurice Pascal Artz: formal analysis (supporting) and investigation (supporting).

Franko Greiner: conceptualization (equal); funding acquisition (lead); methodology (equal); project administration (lead); resources (lead); software (supporting); supervision (lead); and writing—review and editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

REFERENCES


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