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ORIGINAL ARTICLE

Determination of microparticle characteristics in an etching plasma

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The properties of one single melamine formaldehyde microparticle, levitated in the sheath of a low-pressure radio frequency-driven plasma, are determined with a novel procedure using in situ Mie scattering. By placing the measurement equipment inside the vacuum vessel, optical access is not limited to available viewports, and the complete scatter profile can be measured. The developed fitting routine uses peaks and valleys in the angular scatter profile to accurately determine the particle's refractive index, size, and plasma-induced etch rate. In plasma at a pressure of 13 Pa operated at 10 W, obtained refractive indices of 1.657 ± 0.01 are close to those specified by the supplier, while the initial size ($2.168 \pm 0.007 \mu\text{m}$) is relatively 8% off from the supplier-specified value. With an estimated contamination of ambient air of $4 \pm 1\%$ in pristine Ar, etch rates of $1.9 \pm 0.2 \text{ nm/min}$ are measured.

KEYWORDS

complex plasma, etching, microparticle, Mie scattering

1 | INTRODUCTION

Plasmas containing charged macroscopic particles, so-called dusty plasmas, have been topic of research for decades.^[1] Dusty plasmas are found in space as interstellar dust and within the rings of Uranus. Even the early solar nebula appeared to be a dusty plasma.^[2] On Earth, dusty plasmas were first recognized in the semiconductor industry, where particles were spontaneously grown in etching plasmas.^[3] Causing defects on the produced semiconductor structures, these particles were unwanted, and the challenge to prevent their formation and presence was born. Advantageous applications of particle-containing plasmas were identified at a later stage as nanoparticles grown in a plasma can contribute to the production of thin silicon films^[4] and can be used for the production of disperse composite materials.^[5]

The particles in a dusty plasma can be used as probes to study the plasma sheath.^[(6–12)] This plasma sheath is the region at the edge of the plasma where electrons are depleted due to their high mobility, and the remaining ions cause a positive space charge.^[13] Therefore, a potential difference exists between the bulk plasma and the walls confining it, and an electric field points towards the plasma boundaries. The electric field accelerates the ions towards the walls, making the sheath a rather interesting region, not only for fundamental study but also for many applications including etching,^[14] plasma deposition,^[15] and sputtering.^[16]

When immersed in a plasma, particles acquire a permanent negative charge and therefore encounter an electric force in the plasma sheath, next to forces due to, for example, gravity and ion drag. In low-pressure radio-frequent (RF) discharges, nano- to micrometre-sized particles easily find a position where these forces are in equilibrium. Hence, the particle can be confined in the plasma sheath. The equilibrium position can be used to study the force balance on the particle^[17] and determine the electric field of the plasma sheath.^[7]

Because the particle charge and the different forces acting on a plasma confined the particle scale differently with the particles size, a precise determination of this size is inevitable for further understanding of dusty plasmas. Ex situ, the size of particles

is easily determined, for example, by microscopy or transmission electron microscopy.^[18] A popular technique used for in situ size measurements is light scattering. An advantage of this technique is that, as long as the effects of photodetachment are small enough, it has negligible influence on both the plasma and the dust particle and is therefore non-intrusive. Particle sizing on large dust clouds in different plasmas can be performed by light extinction of an incident light beam^[19] or imaging Mie ellipsometry.^[(20–22)] Particle sizing on single particles in an oxygen plasma has been performed by angle-resolved Mie scattering on a small range of scatter angles, with the detection system outside a vacuum vessel.^[23] More recently, angle-resolved Mie scattering with a range of 90° has been utilized to measure the spatially resolved size distribution of microparticles in extended dusty clouds.^[24]

This paper introduces a novel method to measure the size of a single microparticle confined in a plasma. The light scattered from the particle is measured angularly resolved, over an almost complete range of scatter angles. By fitting the local extreme values in the resulting scatter profile, the particle's radius and refractive index are found, even though the particle's size significantly decreases as a function of time during one single measurement. In contrast to ellipsometry, angular-resolved measurements, together with the introduced analysis, output unique values for the particle properties. Compared with the only known angularly resolved scattering measurements at a small range of scatter angles, the large range of measured scatter angles provides the refractive index together with a more accurate particle size.

In this manuscript, firstly, the interaction between microparticles and both the discharge and electromagnetic waves is discussed. Secondly, the conducted research methods are described in the sections Setup, Experiment, and Fitting procedure. Finally, the results are presented, followed by the conclusions.

2 | PLASMA-PARTICLE INTERACTION

Due to the dynamic behaviour of ions and electrons in a plasma, an immersed macroscopic particle interacts with the plasma in various ways. Both the plasma forces acting on microparticles and plasma etching of polymer particles is briefly discussed in the following sections. For a detailed overview of the different forces in a dusty plasma, the reader is referred to Refs. [25, 26].

Particles acquire a net negative charge when immersed in a plasma, and therefore, an electric force is acting on them when electric fields are present. The orbital motion limited (OML) theory is often used to calculate the floating potential of a particle by equating the electron and ion fluxes to the particle.^[27] Considering the capacitance of a spherical particle, the charge is estimated. For micrometre-sized particles and typical plasma parameters the particles may carry thousands of electron charges. Moreover, the charge is proportional to the radius of the particle.

In the plasma sheath (see e.g., Ref. ^[28]), where high electric fields exist, the electric force can be large enough to counter the gravitational force acting on a particle. Therefore, micrometre-sized particles can find a stable equilibrium position and are confined in the sheath. The electric and gravitational forces are the dominant forces acting on the particle for the used configurations. Other forces (e.g., ion drag force, neutral drag force, and thermophoretic force) can be neglected in this work.

Plasma etching is driven by one or more of the following four mechanisms: sputtering by ions, chemical etching by neutrals (including radicals), ion energy-driven etching by neutrals and radicals while ion bombardment chemically activates the etched material, and ion-enhanced inhibitor etching.^[29] In our experimental configuration, the latter option can be disregarded as no inhibitor is present on the dust particles. Chemical etching might take place in the setup due to contamination of the discharge, which acts as a source for etchants like O and OH. Etching by neutrals, or radicals, does not have a preference direction, while sputtering and ion energy-driven etching do have a preference direction. In the latter case, the particle is mainly etched from the discharge side of the particle where the ion impact is higher.

A small model is introduced to understand how the etch rate is dependent on the particle size. Consider the change of the particle's mass M , which is proportional to the cross section σ for the interaction between the particle and the considered species with flux Γ , and the rate coefficient K , which might both be dependent on the ion energy E_i :

$$\frac{dM}{dt} \propto K(E_i)\Gamma\sigma(E_i). \quad (1)$$

When ion energy-driven etching is the dominant mechanism, the etch rate is proportional to the ion flux, which is proportional to the cross section for the ion drag collection force:^[26]

$$\sigma(E_i) \propto \pi a^2 \left(1 - \frac{2e\Phi}{E_i} \right), \quad (2)$$

where a is the particle radius, and Φ is the floating potential of the particle. Now calculating the change in the particles radius, the etch rate gives:

$$\frac{da}{dt} \propto \frac{1}{4\rho} K(E_i)\Gamma \left(1 - \frac{2e\Phi}{E_i} \right), \quad (3)$$

where ρ is the mass density of the particle. This shows that the etch rate is dependent on the particle size when ion energy-driven etching is significant. In the bulk plasma, the particle potential, and thus the cross section, is dependent on the electron and ion temperatures.^[30] In the (pre)sheath, however, the ion current, and hence the particle potential, is also dependent on the local electric field, which accelerates the ions, and on the ion and electron density.^[31] When the particle becomes smaller, the equilibrium position alters to positions closer to the plasma bulk, resulting in a different particle potential.

In case the neutral species flux determines the etch rate, the cross section is not influenced by the floating potential of the particle and is simply given by the particle surface $4\pi a^2$, resulting in a constant etch rate.

3 | MIE THEORY

To enhance the understanding of the performed light scatter measurements, a brief description of the theory of light scattering by small particles is given. For a complete in-depth derivation of the provided theory, see Refs. [32, 33].

Electromagnetic waves that encounter a dielectric material induce oscillating electric dipoles, which in turn radiate electromagnetic waves. The intensity of light scattered from a particle scales linearly with the square of the electromagnetic wave's electric field. Consider a particle situated in the origin. The relation between the electric fields of the incident (E_i) and scattered (E_s) waves with polarisations perpendicular ($\perp$) and parallel ($\parallel$) to the scatter plane are given by

$$\begin{pmatrix} E_{s\parallel} \\ E_{s\perp} \end{pmatrix} = \frac{e^{ik(r-z)}}{-ikr} \begin{pmatrix} S_2 & S_3 \\ S_4 & S_1 \end{pmatrix} \begin{pmatrix} E_{i\parallel} \\ E_{i\perp} \end{pmatrix}, \quad (4)$$

where k is the wavenumber, r is the distance to the origin, and z is needed to correct for the incoming beams travelling over the z -axis. This equation demonstrates that the scattered light behaves like a spherical wave. Of interest are the matrix elements S_1 , S_2 , S_3 , and S_4 , which are dependent on the scatter angle, refractive index, and shape of the particle.

In order to calculate these coefficients and compare them with the measured data, a model is desired. The model to be used in this case is based on the Mie theory, which describes scattering of electromagnetic plane waves on spherical particles. Due to the spherical symmetry of the scatterer, analytical expressions of the scattered fields can be resolved in the complete space as a function of the refractive index of the particle and the particle's size relative to the wavelength of the scattered light.

As spherical particles S_3 and S_4 will vanish, S_1 and S_2 are given by:

$$S_1 = \sum_{n=1}^{\infty} \frac{(2n+1)}{n(n+1)} (a_n \pi_n(\cos \theta) + b_n \tau_n(\cos \theta)), \quad (5a)$$

$$S_2 = \sum_{n=1}^{\infty} \frac{(2n+1)}{n(n+1)} (a_n \tau_n(\cos \theta) + b_n \pi_n(\cos \theta)). \quad (5b)$$

The angle-dependent functions are defined by

$$\pi_n = \frac{P_n^1(\cos \theta)}{\sin \theta}, \quad (6a)$$

$$\tau_n = \frac{dP_n^1(\cos \theta)}{d\theta}, \quad (6b)$$

with P_n^1 being the associated Legendre polynomial of order 1 and degree n . The scatter coefficients a_n and b_n in Equation (5) are

$$a_n = \frac{m^2 j_n(mx) [x j_n(x)]' - j_n(x) [mx j_n(mx)]'}{m^2 j_n(mx) [x h_n(x)]' - h_n(x) [mx j_n(mx)]'}, \quad (7a)$$

$$b_n = \frac{j_n(mx) [x j_n(x)]' - j_n(x) [mx j_n(mx)]'}{j_n(mx) [x h_n(x)]' - h_n(x) [mx j_n(mx)]'}, \quad (7b)$$

with m being the refractive index of the particle relative to the refractive index of the surrounding medium (with refractive index 1) and $x = ak$ being the size parameter found by multiplying the particle size with the wavenumber. Furthermore, there is the Spherical Bessel function of the first kind, j_n , and the spherical Hankel function of the first kind, h_n .

The typical shapes of these scatter profiles are given in Figure 5 later.

FIGURE 1 Schematic view of the setup and cross sections from the (a) top and from the (b) side. (a = vacuum vessel with three viewports; b = plasma; c = microparticle; d = laser; e = polarizer, polarizing the light perpendicularly to the scatter plane; f = laser beam; g = collector rotating around the plasma; h = camera; i = photodiodes; j = pillars between electrodes; k = detection boundaries; l = electrodes; m = stepper motor to rotate the collector around the plasma; n = particle dispenser; o = modification of electrodes for particle confinement; p = spectrometer; q = optical fibre; r = connection to pumping system; s = gas inlet.)

4 | EXPERIMENTAL SETUP

The key features of the used setup are schematically shown in Figure 1. An RF discharge (13.56 MHz, 10 W) is created in argon (13 Pa) between two circular electrodes (140 mm in diameter). The used microparticles are spherically shaped and composed of polymeric melamine formaldehyde (MF).^[34] The supplier specified radius is $2.35 \mu\text{m}$, and the specified refractive index is 1.68 (dispersion relation or imaginary part are not provided by the supplier).^[35] A particle, released from the particle dispenser will find its equilibrium position about 5–7 mm above the electrode, depending on the gas pressure, plasma power, and oxygen content. An indent in the bottom electrode allows for horizontal confinement of the particle.

The incoming light for the scatter experiments is emitted by a cw diode laser (CNI MGL-H-532, 532 nm, <500 mW) and is linearly polarized, perpendicular to the scatter plane. The intensity of the beam is monitored by photodiodes, measuring the intensity of the small fraction of light reflected at the windows before and after the beam has passed through the setup. Light from this beam is scattered by a microparticle and collected by a collector, which couples the light into a spectrometer (Avantes AvaSpec-ULS2048). The collector is placed on a rotating plate so that the scattered light can be measured and angularly resolved. The rotating plate and the collector are placed inside the vacuum chamber because of which the range of scatter angles to measure is not limited to, for example, viewports. The scattered light is only blocked by three electrically insulating pillars that keep the two electrodes at a fixed distance apart. Perpendicular to the laser beam, a camera (ImagingSource DMK 23GM021) is placed to visualize the particle and track its position over time.

The collector has a finite size, and light scattered within a solid angle of 0.002 (corresponding to 3° in the scatter plane) is collected. To fit the data, it is therefore necessary to calculate a numerical integral over the scatter angles within the range of the collector. Not all scatter angles in this detection range lie in the same scatter plane, which is accounted for when comparing the measured data with Mie theory.

FIGURE 2 Video frame of a levitating dust particle above the modified electrode

FIGURE 3 Intensity of light scattered from a dust particle as a function of scatter angle and time. The subsequently measured profiles show that the particle's size is decreasing over time

5 | EXPERIMENTAL PROCEDURE

A picture of the confined particle is shown in Figure 2. Five subsequent Mie profiles of light scattered from this particle are measured. The measurement is performed in a contaminated discharge. Besides argon, water vapour, nitrogen, and oxygen are most likely present in the discharge. The oxygen content is estimated to be $0.7 \pm 0.3\%$ based on the base pressure of the system leading to an estimated contamination of ambient air of $4 \pm 1\%$. The measurement is performed at a gas pressure of 13 Pa and plasma power of 10 W.

The intensity of the scattered light is measured from 5 to 160° with steps of 0.1° . Scatter angles less than 5° are not measured as the spectrometer becomes saturated due to the high intensities of the incoming beam, and scatter angles of more than 160° are avoided so the collector does not block the incoming beam.

After the experiment, the particle is removed from the plasma, and the same measurement is performed with the same plasma parameters and laser power. Subtracting this background from the actual measurement results in the desired scatter profile.

Figure 3 shows the relative intensity of the scattered light from one and the same particle as a function of scatter angle and time. For each scan, a scatter profile is found that is typical for Mie scattering, and the scattered intensity varies with orders of magnitudes for different scatter angles where forward scattering is dominant over other scatter directions.

Looking at the profiles, one observes that the amount of peaks in each scan decreases. This indicates that the particle size is decreasing over time. Making an angular scan takes more than an hour. This long measurement time is needed as (a) a high enough signal should be detected at each measurement angle, and (b) angular resolution should be high enough to be able to perform data analysis. It is very much worth noting that, in this time, the particle size is changing significantly and cannot be assumed to be constant during an angular scan.

Other proof for a decreasing particle size is found in Figure 4. The particle's vertical position, recorded with the camera, is plotted as a function of time. The particle position rises in the sheath during the measurements. Because the forces on the particle scale differ with the particle radius, the equilibrium position is located higher in the sheath when the particle's size is smaller.

A comparison between the experimental data and Mie theory is only justified in the case of spherical particles. Even in the case of ion-assisted etching, causing anisotropy, the particle will retain its spherical shape. It is shown that spherical particles

FIGURE 4 Vertical position of the dust particle above the electrode as function of time. The error bars represent region in vertical direction in which the particle is detected in a video of 4 s

FIGURE 5 Two theoretical Mie profiles with a decreasing radius. For both profiles, the particle radius is decreasing linearly over time and thus as a function of scatter angle. The radius at 0° is $2.0 \mu\text{m}$ for both profiles, the line profile has an etch rate of 0.6 nm/min , and the dotted line has an etch rate of 1.2 nm/min

with small inhomogeneities will encounter a net angular momentum from the ion flux, making the particle spin.^[36] Furthermore, due to the possible loss of symmetry, an electric dipole moment is induced, and the longest dimension of the particle will align with the electric field. These two effects will continuously change the orientation of the particle, and the particle is etched isotropically over time, independent of the etch mechanism.

It is assumed that during one scan, the etch rate is constant. A different particle position might change the etch rate, but results show that, within one scan, the assumption of a constant etch rate is valid.

6 | FITTING PROCEDURE

A novel fitting procedure is developed that uses the particle's scatter profile to deduce particle characteristics. By fitting the measured data with Mie theory, the refractive index and particle size over time can be retrieved. The particle size is assumed to decrease linearly over time during one scan over all scatter angles. So, the particle's size as function of time is fixed by two values, the etch rate and the initial particle size.

The shape of the scatter profiles is determined by the amplitude functions S_1 and S_2 , which depend on the refractive index, particle radius, and the scatter angle. In the research presented here, light is perpendicularly polarized with respect to the scatter plane. Therefore, to find the particle properties, the measured scatter profiles should be compared to S_1 only.

Using a conventional least square method is not sufficient to fit the data. Two fit parameters that lag physical interest need to be introduced: one scaling parameter, because the intensity of the light is measured relatively, and an offset parameter, which is important because the intensity of the scattered light varies by orders of magnitudes during one scan. The error landscape would be a five-dimensional space containing many local minima, and finding the global minimum would be a matter of trial and error.

The way to fit the data and avoid the above-mentioned difficulties is by looking at the local extreme values in the data and comparing these with the local extreme values predicted by Mie theory. The position (in terms of scatter angle) of these peaks and valleys are distinctive features of the scatter pattern and are independent of scaling factors or offsets. In addition, the computing time decreases significantly as not all data points, but only the position of the local extreme values, are needed in the calculation.

For illustration, Figure 5 shows two theoretical scatter profiles where the intensity is given as a function of scatter angle and time. At small scatter angles, the radius is the same for both profiles, but as they are chosen to be subject to different etch rates, the particle size changes with time. Hence, there is a difference between the positions of the peaks at larger scatter angles.

FIGURE 6 Fits for subsequent scans of the scattered light on the same particle. The grey indicators mark the peaks and valleys that are included in the fitting process

To fit the measured data, the difference between the measured profiles and the fit should be minimized. For each guessed fit, the extreme values are calculated. After that, for each measured extreme value, the distance to the closest extreme in this fit is given by δ_i (two examples are indicated in Figure 5). To find the fit parameters,

$$\Delta = \sum_i |\delta_i| \quad (8)$$

is minimized so that the peaks and valleys are in the same position.

In order to make this method work, the correct positions of extreme values in the scatter profile are necessary. These points are obtained by fitting a quadratic function through the data points that surround the peaks and valleys. The position of the

TABLE 1 The found fit parameters for each scan together with corresponding error

#	m	a_0 (μm)	$\frac{da}{dt}$ (nm/min)	$\frac{\Delta}{N}$ ($^\circ$)
1	$1.651-0.001i \pm 0.003$	2.168 ± 0.007	1.9 ± 0.2	0.4
2	1.655 ± 0.009	2.028 ± 0.004	1.9 ± 0.1	0.5
3	1.665 ± 0.009	1.891 ± 0.008	2.1 ± 0.2	0.8
4	1.651 ± 0.002	1.768 ± 0.005	2.0 ± 0.1	0.5
5	$1.661-0.002i \pm 0.01$	1.629 ± 0.006	1.8 ± 0.2	0.5

FIGURE 7 Fitted particle size as function of time. The black line is the values used for the fit, and the grey areas show the error margin found for each fit

extreme value of this quadratic fit is then used as the position of the peak or valley. To reduce the influence of noise on the results, only the clearly detectable extreme values should be included in the fit algorithm. The candidate extreme values are filtered using some rules. The exact procedure to determine the representative extreme value and the corresponding position is given in Section 8.

7 | RESULTS

During five subsequent scans, the angular scatter profiles are fitted with the procedure described above, as shown in Figure 6. Note that the total measurement time for this single particle is almost 6 h. Grey circles mark the used peaks and valleys. The grey areas in Figure 6 mark scatter angles where the light is blocked by one of the pillars in the setup.

The etch rate is fixed while fitting each individual scan (it may be different for subsequent scans). The range of the used fit parameters, which are the same for all scans, are taken sufficiently widely around the expected values given by the supplier (1.62–1.74 for the refractive index, 1.2–2.8 μm for the particle size). With these rather wide parameter ranges, data analysis typically takes 3 hr per fit.

For each of the five scans, a refractive index m , etch rate da/dt , and initial radius a_0 are found by fitting the data. The obtained values are presented in Table 1, together with the corresponding errors Δ relative to the amount of used extreme values N . The fits result in a particle radius as a function of time, which is shown in Figure 7 for all fits. Although the five consecutive fits are made completely independently, the particle radii are in accordance with each other and are decreasing linearly in time, that is, for each fit, the particle radius at the beginning of the scan is close to the particle radius at the end of the previous scan. Combining the results, a refractive index of 1.657 ± 0.01 and an etch rate of 1.9 ± 0.2 nm/min is found. The etch rates are the same, and values for the refractive index are consistent with each other, although they are slightly different from the value given by the supplier.

The error in particle size is estimated by comparing two calculated profiles with a different radius. The difference in radius between the two calculated profiles will result in an error Δ_r (similar to Equation 8). Increasing the difference in radius will push the positions of the peaks further apart and thus will increase Δ_r as well. The estimated error in particle radius is defined as the difference between the radii of the two calculated profiles, for which Δ_r is equal to the error of the fit Δ , both relative to the amount of extremes that is used.

The error estimation for each scan is shown by the grey areas in Figure 7. The error in the refractive index is estimated in a similar way, looking at the difference in refractive index instead of the difference in particle radius. The error in the etch rate is the minimal and maximal etch rates, for which the particle radius stays within the error margins of the radius. The error estimated in this way for the different fit parameters estimated with this procedure is the error from the fit algorithm.

As mentioned in Section 2, the etch rate of a particle may be dependent on the position in the sheath. For different sheath positions, the density of etchant species might be different, as is the ion energy and flux. Although the position of the particles presented here changed during measurements, the etch rate is found to be constant over time. This indicates that the spatial dependence of the etch rate is small enough and can be neglected.

A special note must be made of the complex part of the refractive index κ , that is, the extinction coefficient defined by $m = m_{\text{Re}} - i\kappa$, which is unknown for MF particles. In a first fit, κ is assumed to be zero. After that, the fitting algorithm is used once more, where the extinction coefficient is changed from 0 to 0.01 in steps of 0.001. A non-real value for the refractive index results in slightly different values for the other fit parameters. For all fit parameters, this difference is much smaller than the error estimates. The exception is for the particle size at the start of the measurement, but this difference is still less than 1%. This justifies the assumption of a real refractive index for the MF particles.

8 | CONCLUSIONS

The properties of spherical particles, levitated in the plasma sheath, are determined by angularly measuring the light scattered from this particle and fitting these data with Mie theory. A novel fitting method is introduced, which fits the local extreme values of the measurement with theory. This method has the advantage that not every data point is used for fitting, and only essential fit parameters that are physically interesting are used.

With the used technique, it is possible to determine the refractive index and particle size extremely accurately over a long period of time, even when the particle becomes smaller by plasma etching during the measurement. Assuming the extinction coefficient to be zero results in a good approximation of the particle properties. The assumption of a constant etch rate is justified because the particle size decreases almost linearly over a time span of almost 6 hr, which is verified by comparing the consecutive independent fits.

The refractive indices obtained in the different intervals are consistent with each other, as are the found particle sizes. The small differences in particle size at the start of a new fit are consistent with the small differences in the refractive index found and are expected to be related.

The difference between the refractive index and particle size with the data provided by the supplier is a surprise. The values for the refractive index are between 1.63 and 1.67, which is lower than the specified 1.68. Conditions under which the supplied refractive index is determined are unknown; besides that, deviation from the specified refractive index might be caused by an increased particle temperature or outgassing of the microparticle in the vacuum chamber.^[37]

The deviation of the measured particle radius at the start of the measurements ($2.168 \pm 0.007 \mu\text{m}$) with the one specified ($2.35 \pm 0.04 \mu\text{m}$) is remarkable. The initial particle size is found to be a considerable 6–13% smaller than provided by the supplier. The particles are confined in the discharge for a few minutes before the measurements. Considering the etch rates, this short period cannot be held responsible for the difference in the particle sizes.

The consequences of assuming the wrong particle size are not to be underestimated. The particles used in this research are commonly used as probes to investigate the plasma sheath.^[(7,31,12)] Gravity, particle charge, and ion drag forces are dependent on particle size and are used as an intermediate step to determine plasma parameters, if not the subject of investigation itself. Knowledge of the correct particle size is crucial for this kind of research.

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APPENDIX

To determine the correct fit parameters, it is important to find the correct position of the extreme values in the measured scatter profile. Looking for minima and maxima in the data, several peaks and valleys are caused by noise. These extreme values should not be used in the fitting algorithm as there is no corresponding extreme value in the theoretical Mie profile. In addition, noise might cause the position of a useful peak or valley to be determined incorrectly. These issues can be avoided by applying some criteria to the candidate extreme values.

The data values of the measured profile vary with orders of magnitudes. To still be able to apply the same criteria to both the peaks and valleys, the natural logarithm of the data is used to determine their positions. After that, a smoothing algorithm is performed over the measured profile. This means that, for each data point, an average over the nine nearest data points is calculated. Here, the noise is reduced while the position of the important extreme values is not influenced.

When searching for peaks and valleys in this smoothed data, it is necessary that the distance between two peaks and the distance between two valleys is at least 5° . If multiple peaks are found within this range, the highest peak is used. In addition, the found extreme values should have a minimum prominence^[38] of 0.2. To determine the position of the peaks and valleys more correctly, the nine data points nearest to the found extreme value are fitted with a parabola. This parabola gives a more accurate position as more data points are used to calculate it. The found points are only included in the calculations if the R^2 value of the parabolic fit is at least 0.6. Note that these values are applied to the non-normalized data.

The consequence of taking these steps to find the peak position is that the peaks are found on the expected positions in the measured profile. Without the mentioned restrictions, peaks that arise due to noise are included in the fit algorithm. Hence, incorrect fit results would be obtained.