

Reply to RC1:

The authors express their gratitude to Referee 1 for dedicating time to reviewing the manuscript and providing concise comments and questions. We have thoroughly addressed each of the points raised and indicated changes made in the manuscript by highlighting them in the text. Below are the referee's comments in black text followed by our replies in blue text.

1. What is the driver voltage amplitude and how is the appropriate magnitude of this chosen?

The magnitude of the applied AC-voltage was kept below 1.5kV peak-to-peak in order to make sure that no electrical discharges occur within the EDB. Due to the application of two independent AC-potentials, i.e. the levitation and the excitation potentials, potential differences equivalent to the sum of both amplitudes will occur. The operational amplifier was programmed in such a way as to compensate the nonlinear output of the transformers. In this way it was possible to maintain a constant output amplitude across the entire frequency range. We added this information to the manuscript text on page 7 as well as to the SI in section 2.

2. What is the uncertainty in the particle size, and how much of this translates to uncertainty in viscosity and surface tension? Equations 6 and 7 both show dependencies on R^2 or higher, indicating some significant propagation of error. The authors chose to use the phase function approach, which for large particles can yield reasonable accuracy when comparing measured spectra to Mie theory simulations. There are several approaches to sizing using these data, as described in the SI. Thus, it would be convenient to describe how the angular scattering pattern is used in the main text along with the estimated uncertainty.

As outlined in Section 3 of the SI, the relative uncertainty in particle size is contingent upon whether single images (4% uncertainty) or a filtered set of multiple images (1% uncertainty) were utilized for analysis, with the resulting $R(t)$ data being effectively smoothed through the application of an evaporation model. The uncertainty in droplet size significantly contributes to the overall uncertainty in viscosity and surface tension and improving the size determination method would provide great benefit to a future iteration of the method. Because of its significance, we added a short description of the size determination method to the main text on pages 6-7, while keeping the detailed description in Sect. 3 of the SI.

3. I would suspect that when the droplet is driven at a frequency resonant with a mode of oscillation that the amplitude of the oscillation would increase significantly, leading to a higher scattered signal from the droplet. Using a dual-phase lock-in, the amplitude of the scattering and the phase of the signal, both as a function of frequency, could be analyzed as the driver frequency is swept. Has amplitude information been measured and/or analyzed to identify the resonant frequency? This would presumably directly yield ω_0 for an underdamped oscillation, but perhaps not yield sufficient information on the damping constant for viscosity analysis.

We determined ω_0 and the damping constant γ by analysing the oscillation amplitude. However, we found that amplitude analysis was associated with a significantly greater uncertainty compared to phase analysis and that it is only reasonably effective for non-evaporating or extremely slowly

evaporating droplets ($dr/dt \ll 10 \text{ nm/sweep}$). The reason for this lies in the highly nonlinear characteristics of the measurement signal, specifically in how the intensity of scattered light varies with the oscillation coordinate.

To obtain absolute values for the amplitude of oscillation (i.e. amplitude of actual droplet deformation), it would be necessary to develop an additional analysis algorithm. For the purpose of testing, we manually compared measured and simulated light scattering time series to obtain droplet deformation amplitudes. The corresponding result has been included in panel B of Fig S11 in the SI. However, this demanded a considerable investment of manual analysis effort, which we found to be unjustified when contrasted with the straightforward and relatively accurate results achieved through phase analysis.

These factors led us to focus on the phase analysis for our methodology.

4. What are the limits of viscosity that can be probed with this method. The viscosity values reported here are all many orders of magnitude lower than what is typically considered to be a viscous particle. Is there a limit on the viscosity that can be measured due to the limited response of a more viscous particle to any kind of shape deformation? Is this connected with the deviation of the measure viscosity in this approach from the literature range (Figure 3 for example)?

In our approach, we employ a phase-sensitive detection (PSD) technique. Fundamentally, PSD proves to be highly effective in extracting minute signals concealed amidst significant background noise levels. As a result, the amplitude of the signal and the extent of the oscillation that can be induced are unlikely to pose concerns using this methodology.

However, there might be other factors limiting the viscosity range in our method, such as obtaining a sufficient amount of data to precisely determine the natural frequency of the undamped oscillation ω_0 and the damping constant γ from the theoretical fitting process of the phase-frequency data. Ideally, this entails carefully selecting the frequency sweep range, encompassing ω_0 where the phase shift reaches $\pi/2$, and encompassing a sufficiently broad span of both lower and higher frequencies. This is particularly effective for underdamped oscillations. However, for overdamped oscillations, the amplitude-frequency curve and the phase-frequency curve adopt a relatively flat profile, which indicates a diminished information embedded within these curves. Consequently, the accurate extraction of ω_0 and γ using our methodology can become challenging under such circumstances. Based on this reasoning, we estimate an upper limit for the viscosity measurement in the range of a few Pa s, while the determination of a lower limit remains elusive. We have added this statement to the main text on page 13. The detailed limits of viscosity will be explored in further measurements.

5. Were measurements performed on particles that spanned a range of charge states? When calculating the charge according to the SI, no gas flow drag factor is reported. How does the gas flow in the chamber affect the force balance? How big is the uncertainty in the charge and how much does this contribute to the calculation of surface tension and viscosity?

The span of droplet charge states measured in this study was between 0.67 pC and 0.84 pC. We estimated the relative uncertainty in droplet charge measurement to be about 3.5%. We added this information to the SI on page 5 and a statement in the main text referring to the SI on page 9. A systematic study of the influence of droplet charge on this method was not performed.

Equation (6) in the manuscript indicates that the viscosity is independent of the droplet charge. However, following Eq. (7) the surface tension is dependent on Q . To evaluate a potential influence of the charge on this method it is convenient to rewrite Eq. (7) as:

$$1 = \frac{\rho\omega_0^2 R^3}{4\sigma} + X,$$

where $X = Q^2/Q_R^2$, the fissility parameter, with droplet charge Q and Rayleigh charge Q_R . From Eq. (7) in the above form it becomes apparent, that the contribution due to the term containing the droplet charge is small, as long as the fissility is much smaller than 1. For the droplets under investigation in this study, the fissility is typically below $X=0.01$ upon injection and never exceeds $X=0.1$ throughout the measurement duration. Consequently, the uncertainty in the charge does not contribute significantly to the uncertainty in the surface tension. Instead, it is the power dependence on the droplet radius, that emerges as the primary contributing factor.

In addition to this analysis, the uncertainties of dependent variables are consistently calculated following the principles of error propagation.

Regarding the gas flow, the EDB was operated without any gas flow, eliminating the need to account for drag force in the force balance. Gas flows through the EDB have been utilised in previous experiments in our group. In order to avoid ambiguity with these experiments, we added the above information to the main text on page 5.

6. What is the timescale for evaporation in the measurements shown in Figure 6? Does the decrease in surface tension arise from the adsorption of known contaminants from an external source (i.e. are there lubricant oils that might be outgassing, low purity gas cylinders, etc.), or are these present in the particle and become more concentrated at the surface as evaporation occurs?

Figure 6 in the manuscript shows surface tension measurements for five individual droplets (1 water and 4 sucrose solution droplets) while evaporating in the EDB. Each of these measurements was recorded over a time span ranging from 100 to 120s. We added this information to the main text on page 12.

The decrease in surface tension could arise from adsorption of contaminants or those that are already present in the solutions studied. However, it is difficult to say for certain without further investigation. Special care was not taken to prepare solutions in an ultra-clean environment. As stated in the response to comment 5, no gas flows were used during the experiments. However, in the event that discharges occurred, the EDB was purged with laboratory grade synthetic air which is prepared on site from liquidised nitrogen and oxygen. Low purity gas cylinders and lubricant oils were not utilized. As in previous experiments in our group involving EDBs great care was taken to keep the EDB in a clean condition.

7. How large are the oscillations in the particle shape?

Obtaining the oscillation amplitude from the measurement is challenging due to the complex nature of the time dependent light scattering signal. See also our reply to comment 3. Nevertheless, by manual analysis of the signal we found that oscillation amplitudes as small as a few Angstroms could be analysed.

This is further supported by theoretical evaluation of Eq. (1) from the manuscript. The theory shows that the oscillation amplitude of the equatorial radius of a water droplet at 0°C ($R=50\mu\text{m}$, $Q=0.8\text{pC}$) reaches a value of 1.5nm off-resonance (5kHz, 1kVpp) and up to 12nm at the resonance (11kHz, 1kVpp). To provide the reader with an understanding of the estimated range of the oscillation amplitude, we have included a statement in the main text that presents our estimate on page 8.

8. What size range of particles can be explored using this technique? Being able to access particles that span a wide range of surface-volume ratios would facilitate an exploration of surface partitioning and the influence of surfactant depletion etc.

We estimate that for our experimental setup the range in droplet diameter is limited between 5 to 200 μm . Please refer to our [reply to comment CC1](#) for further information. We added a statement on the size limitations of this setup to the manuscript on page 6.

For this technique it may turn out to be challenging to overcome these limits whilst simultaneously utilizing electrodynamic levitation and electrodynamic excitation. However, the combination of acoustic or optical levitation with electrodynamic excitation could potentially open up the possibility of accessing a broader range of droplet sizes.

Further changes made to the manuscript:

Page 1, line 14; Page 13, line 337

Based on our now refined assessment of the range limits of the viscosity we removed the inclusion of “within the semi-solid range” from the abstract and the discussion.

Page 3, line 95

We removed the sentence: “Currently, viscosity values for dilute droplets are derived from models.” This sentence was a remnant from a previous version of the manuscript among the co-authors, and we believe it is not relevant anymore in the context of this paragraph.

Page 5, line 152:

We added information on the ambient conditions surrounding the levitated droplet.

Page 6, line 176 – page 7, line 180

We added a statement concerning the consideration of the evaporative cooling effect on the droplet temperature.

Further changes made to the SI:

- A typo was corrected in equation E1
- A typo was corrected in the EDB geometry constant stated on page 5 below equation E1