Reply to RC2:

The authors extend their appreciation to Referee 2 for reviewing the manuscript and providing concise comments and questions which also helped us to locate two typos in an equation in the manuscript and SI. We have thoroughly addressed each of the points raised and indicated changes made in the manuscript by marking them in the text. Below are the referee's comments and questions in black followed by our replies in blue text.

1. The only comment I would like to ask the authors to consider is that they explain/estimate in more detail the limitations of the technique in both upper viscosity values and range of surface tension are.

We now elaborate in more detail on the limitations of the technique with respect to the range in viscosity and surface tension. We have included relevant statements in the manuscript on pages 13-14.

2. I was particularly interested in understanding the upper limit for the viscosity. In order to gain a better feeling for the behavior of the phase shift, I put some numbers into eq. (7) to estimate the natural angular frequency, ω0, namely (R=50 µm, σ=70 mN/m, ρ=1000 kg/m3, Q= 0.8 pC) and came up with 47 kHz. Using eq. (6) with a viscosity of 2 mPa s, I calculate a γ of roughly 4000. Putting those numbers into eq. (5), I cannot reproduce something similar to what is shown in Fig. 2. Most likely this is a mistake on my side, but the authors could provide in the SI some numbers on, ω0 and provide a plot where they show the expected phase shift assuming log spaced viscosity data keeping all other parameters constant.

We cannot say for certain what caused the discrepancy between this calculation and our results. In order to facilitate the comparison, we included the droplet charge, as well as results from the fitting procedure in the caption of Fig. 2. We have added an additional section to the SI, Section 6, where we show the expected phase shift as well as the amplitude of oscillation for a range of droplet viscosities. We anticipate that this will help in reproducing our results.

Initiated by the referee's comment we also checked our equations and calculations. While doing so we noticed a typo in eq. (5) in the manuscript which might be contributing to the above discrepancy. We corrected the equation in the revised manuscript. The typo does not impact the results presented in the manuscript as in our analysis the correct equation was utilized.

Furthermore, the value R=43.2µm originally cited in the manuscript as the droplet radius in the text above and in the caption of Fig. 2 does not actually correspond to the droplet radius. Instead, it corresponds to the time in seconds when the sweep transverses ω_0. The accurate droplet radius at this moment was R=44.3µm according to $R^2(t) = R_0^2 - b \cdot t$ with $R_0 = 50\ \mu m$ and $b = 12.48\ \mu m^2 s^{-1}$ as given in the caption to Fig. S3 of the SI and using $t = 43.2\ s$. Incidentally, both numbers are very similar and the incorrect number was inadvertently entered into the manuscript. We have replaced the number with the correct droplet radius.

Regarding the last mentioned point of the referee's comment, ω_0 is the frequency of oscillation of the undamped oscillator. This parameter is therefore by definition independent of viscosity. It is a noteworthy and interesting feature of the damped driven oscillator, that the frequency of the undamped system still appears in the equations of the damped system. The position of 90° in the phase-frequency curve which indicates $\omega = \omega_0$ consequently is invariant to the viscosity of the droplet. This property might be considered as an advantage of analysing the phase instead of the amplitude response for some mildly overdamped systems.

3. Fig. S4: Please explain what is causing the apparent decrease in droplet charge after 70 s in a bit more detail. What is the size of the particle at this time of evaporation? Is this reaching the stability limit of the EDB or is the DC-feedback loop to keep the droplet in the center of the EDB no longer working?

The size of the droplet can be taken from Fig. S3 as both data series were recorded from the same levitated droplet. We included this information in the figure caption of Fig. S4 of the SI.

Deviations in the charge data like the one presented in Fig. S4 are usually not connected to the stability limit of the EDB as both occur independently. In the example shown in Figures S3 and S4, the time series ends at 160s because the droplet reached the stability limit for the chosen fixed EDB settings. However, the deviation in the charge curve occurred significantly before.

To explore the origin of the deviation in the charge curve is outside the scope of this paper. However, we will provide a more detailed explanation below which hopefully sheds some more light on this.

The charge curve shown in Fig. S4 was calculated using Equation E1 which in turn was derived under the assumption that the droplet resides in the electrodynamic centre of the EDB, i.e. at z=r=0. This is the singular location in the EDB where the electric force due to the AC levitation potential (the pseudopotential) vanishes and only gravity and static DC electric force act on the droplet. Outside the EDB centre the average electric force due to the AC-levitation field has to be taken into account in the force balance. If the droplet is not positioned in the EDB centre, application of equation E1 results in deviations from the constant charge as visible in Fig. S4. Based on the graph, we make an estimation that places the droplet's centre of mass approximately 5µm above the EDB centre. This assessment relies on treating the droplet's charge as a point charge, as opposed to a surface charge. Consequently, we recommend to exercise caution with this estimation.

4. Connected to the data shown in Fig. S4: How is the flow in the EDB affecting the determination of Q based on the applied DC-field? Does the drag force cause a systematic uncertainty here?

The EDB was operated without airflow during our experiments. Airflow was employed only for purging purposes between experiments. We included a statement in the manuscript to avoid ambiguity with previous studies made in our group where airflows have been utilized simultaneous to droplets being levitated. The statement was inserted on page 5.

We note, that if the flow conditions are well known, the drag force can be included in the force balance. Much like in Millikan’s famous experiment, the air flow does not hinder determining the droplet charge as long as the flow conditions or the relative velocity of the particle in the air are well known.

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