Reply to comments on "Technical note: Characterization of a single-beam gradient force aerosol optical tweezer for droplet trapping, phase transitions monitoring, and morphology studies" by Xiangyu Pei et al.

Reply to Anonymous Referee #2

The authors systematically introduced their aerosol optical tweezers coupled with Raman spectroscopy system in a detailed way. They developed a new environmental chamber which has a double-floor and a replaceable intermediate plate, which facilitates stable trapping while introducing different species such as VOC and SOA to the system. In addition to studying the liquid-liquid phase separation (LLPS) of premixed NaCl and 3-methyl glutaric acid (3-MGA) droplets, they were able to in-situ generate and deposit SOA on a NaCl microdroplet and investigate LLPS afterward. Their findings indicate that the AOT system is a powerful tool to in-situ monitor changes in physical and chemical properties of droplets during their evolution under different conditions. I would recommend the paper for publication after addressing the following questions.

Response: We truly appreciate the constructive comments by the referee. These comments are valuable and helpful for improving the quality of our paper. Below we provide a point-by-point response to each comment. The responses are shown in brown and bold fonts, and the added/rewritten parts are presented in blue and bold fonts.

Major comment:

The RH is a key parameter in the LLPS measurements, and the authors compared their experimental measurements with theoretical calculations for the relationship between wet particle diameter and RH (Figure 4a). They only provided the data for a short RH range (around 77% to 86%), and it appears that that experiment deviated from theory at higher RH (83%-86% RH). What are the results for a higher RH range, say 86% to 99%?

Response: Thank you for the question. In Figure 4(a), the RH was measured with the RH sensor at the exit of the chamber, while wet particle diameters were retrieved from the spectra with the Mie fitting program MRFIT. The typical and maximal tolerances for the RH sensor are ±1.5% and ±1.8% when RH is lower than 80%, respectively. When RH increases from 80% to 100%, the typical and maximal tolerances for the RH sensor also increase linearly until ±2% and ±3%, respectively (Sensirion SHT85 datasheet). As a result, the measured RH values at high RH have more uncertainty. In Figure 4a, we think that the experiment deviation from theory is due to the increasing measurement uncertainty of the RH sensor. When RH is higher than 85% during this experiment, the wet particle diameters retrived from the program MRFIT represents hysteresis. For example, when RH is about 86%, the wet particle diameters are in the range of 11.8-12.2 μm, and the range (0.4 μm) is much larger than the value (~0.01 μm) when RH is lower than 86%, which is shown in Figure 4(b). This hysteresis can be explained by that when RH is high, more WGMs peaks represent in each spectrum, the assignment of WGMs labels may have differences since different combination of WGMs labels can lead to the same WGM positions. Considering all the factors above, we still think that the measured values and theoretical values are consistent.

Minor comments:

OT was introduced as the abbreviation of optical tweezer in Line 46, there is no need to introduce it once more in Line 51.

Response: It has been changed accordingly.

Line 55: Change "This results" to this "This result".

Response: Thanks for the comment. It has been changed accordingly.

"This result in a significant enhancement of stimulated Raman scattering signals at specific wavelengths, which are referred to as whispering gallery modes (WGM)."

Line140: "…to prevent droplet deposition" the surfactant cannot prevent droplet deposition. It helps the deposited droplets to spread on the surface of the coverslip.

Response: Thanks for the comment. The sentence has been modified into:

"Inside the lower part, a glass coverslip is placed and soaked in a surfactant solution (a 50:50 water-to-Decon 90 solution). The surfactant solution is used to help the deposited droplets to spread on the surface of the coverslip."

Line 183: developed by, something is missing after "by".

Response: Sorry for the missed information. The sentence has been modified into:

“To address this issue and retrieve the diameters and refractive indices for core-shell droplets, we employ another program called Mie Resonance Shell Fitting (MRSFIT), developed by Vennes and Preston (2019).”

Reference

Sensirion SHT 85 datasheet, https://sensirion.com/media/documents/4B40CEF3/61642381/Sensirion_Humidity_Sensors_SHT85_Datasheet.pdf