Manuscript Review

Application of linear polarized light for the discrimination of frozen and liquid droplets in ice nucleation experiments

The study that is presented in this manuscript describes an application of polarized light measurements for discriminating water droplets from ice crystals for the purpose of ice nucleation experiments. The abstract was intriguing when it suggests a new approach for interpreting the measured, polarized component of scattered light, i.e. the pulse width. Unfortunately, this is not presented with a convincing argument and, in general, the overall paper is a disappointment, in my opinion. Had I been given the option of reviewing this manuscript prior to its publication as a discussion paper, I would have recommended that it be returned to the authors for further work. In my opinion it is much too premature to be published in its present form and I cannot recommend its publication in AMT in anything close to its present form.

There are multiple issues, that I will address in detail below, that prevent this study from making a useful contribution to measurement science. Broad statements are made with no concrete evidence or references about various aspects of the measurements system that are critical to the reader's understanding of its underlying performance. The numerical simulations are performed, and conclusions drawn assuming an ensemble of randomly oriented spheroids yet the instrument described does not measure ensembles of particles but measures particle by particle. Hence, the whole theoretical basis is suspect from the very beginning. The system is designed to differentiate water droplets from ice crystals on a particle by particle basis, not from an ensemble like lidar systems do. This means that the authors are not taking into account the fraction of ice crystals that will have an orientation such that they appear as spheroids, i.e. they never discuss the uncertainty in their derived ice fraction due to orientation.

The technique that they propose to differentiate water from ice based upon the pulse width was, to this reviewer, incomprehensible with respect to why ice crystals would have a distinctively different height to width relationship than water droplets. They show one set of frequency histograms that are supposed to convince the reader that ice crystals have a pulse width distribution different than water droplets with no theoretical basis. They show a highly simplified Gaussian curve that is supposed to represent how the pulse shape looks as a particle passes through the beam; however, even though they are digitizing at a high rate, and could show actual pulse shapes for water droplets and ice crystals, they never do. In addition, their theoretical curve of pulse height versus pulse width is far from what is actually measured, yet they use this relationship as a definitive way to separate water from ice.

In my opinion, the geometry of the optics offers a really substantive way to separate water droplets from ice crystals given the two planes of detection, yet this is not even considered. Other studies have clearly shown that a comparison of two components of the polarized, scattered light can differentiate spherical from aspherical particles, e.g. Nicolet et al., 2012 and Glen and Brooks, 2012; however, not only do the authors not look at these relationships, they don't even reference the studies that have shown that it works.

It is quite possible that the technique has something unique to offer the measurement community but the results that have been presented here are unconvincing and I would ver wary of any ice fractions that are reported from this instrument using the proposed technique.

Specifics

Abstract: Line 4. "…measurements of the depolarized component of light…". No, single particles do not depolarize incident light. They change the state of depolarization. The scattered light remains polarized but instead of a single plane of polarization there are two at right angles so that the light is now elliptically polarized. See Harris-Hobbs and Cooper, 1987, in their appendix they explain the inappropriate use of the term "depolarization". Only when light is scattered by a large ensemble of particles will the resulting scattered light have random polarizations, i.e. depolarized.

Page 5747: Line 5. Some of the most important references on measurements of polarized light are missing here, i.e. Fukuta amd Kramer (1968) who were the first to use polarized light to detect ice crystals in an IN chamber, Nicolet et al. l 2012 who did extensive simulations of light scattering from spheroids in different orientations (more on that below) and Glen and Brooks, 2012 who show that not only can aspherical particles be clearly differentiated from spherical, but that the backscattering signal is quite strong enough to get useful information, contrary to what the authors in the current paper state several times with no justification whatsoever.

Page 5747: Line 11. "….to avoid complication…". This is the first of several occasion swhere it is stated that backscattering measurements are complicated. Baumgardner et al, 2001 first showed that backscatter as one component of the CAS measurement system works quite well and the many CAS units that are currently in operation belie the assertion in this paper that backscattering is difficult and that such signals are too weak to be useful. Glen and Brooks, 2012 also show the error in statements of this type.

Page 5748: Line 14. "…operated in the immersion freezing mode..". Although it is not the intent of the manuscript to discuss the LACIS, per se, I am curious how the immersion mode of nucleation can be differentiated from contact mode nucleation in this flow tube configuration with its miniature cross section.

Page 5759: Line 25. The actual sample volume is never shown, i.e. it is supposedly defined by the intersecting fields of view, but this means that particles will be passing through regions of varying intensity, given the Gaussian distribution of the laser beam. This region can be defined precisely using ray tracing and validated with a droplet stream, such as shown by Lance et al. (2010) who validated the theoretical sample area of an optical particle spectrometer by mapping with a droplet stream. The pin hole does not replicate scattering by single particles as it diffracts and does not refract light. In addition, even though it is an approximation, the results are never shown of this mapping so the reader is never aware of just how serious the edge effect issue is.

Page 5760: Line 9: To what sizes was the TOPS calibrated? Over the entire size range? And what is the size range of the TOPS?

Page 5760: Figure 4 and all other figure showing Size Parameter. The actual optical diameter should be used, not the size parameter so that the reader can actually see the regions of uncertainty in terms of actual size. The authors state maximum uncertainty of 0.5 μm but I think that in some regions that uncertainty is much larger.

Page 5760: Theory Section. In my opinion, this section is much too large. Most of the theoretical derivations and equations are straight out of light scattering text books or have already been recently described in detail by Nicolet et al., (2012) or Schnaiter et al., (2012). Not only is this amount of detail not needed, in the end isn't actually relevant to the way that the TOPS is eventually used try and differentiate liquid from ice. In addition, as discussed in my opening comments, the T-matrix code is used incorrectly to calculate average values of randomly oriented spheroid, unlike the calculations by Nicolet et al (2012) who show quite clearly why you have to do the calculations for many different orientations to underscore how important orientation is when measuring particle by particle and estimating uncertainties in estimated ice fractions.

Page 5763: Line 27. "Calculations of the response functions of spheroid particles in all *fixed* orientations are too time consuming…" This is a very weak and unconvincing argument given that Nicolet et al. have done this, as have others. It is exactly this type of simulation that is needed to understand the system performance.

Page 5765: Line 1. Why are pulse shapes never actually shown from the three detectors under conditions of droplets or ice crystals? With no comparisons with the idealized wave forms that are shown, the reader cannot be convinced that this is anything other than an intellectual exercise, especially since the following derived equation does not fit the actual measurements very well at all, given that the Y-axis is logarithmic.

Page 5765: Line 17. As previously mentioned, the pin hole does not simulate particle scattering. A spherical droplet would have isotropic scattering so that all three detectors should be viewing the same solid angle and intensity of scattered light. The scattered light from the pin hole does not scatter isotropically. Furthermore, the results from this mapping are not shown. Why not?

Page 5766: Line 9. The problem with this derivation is that it does not account for the fact that a larger particle exceeds the threshold sooner than the smaller because when it is only partially into the beam it scatters enough light to exceed the threshold whereas the smaller particle must penetrate farther. In addition, this derivation assumes that the particles are passing directly through the center of the Gaussian beam; however, there is a distribution of shapes as particles with the same size pass off center and hence have a difference height to width relationship. This is clearly shown in Fig. 7 where the pulse heights of the smaller particles with shorter pulse widths are overestimated by the theory while larger particles are underestimated.

Page 5767: Line 1. “The measured curve shows a general agreement…”. This is overstating the fidelity of the agreement. A error calculation would show that the agreement is actually not that good.

Page 5767: Line 10. “…broadening of the droplet mode and a more compact ice mode”. This is stated with no obvious explanation or evidence. I could not understand what this meant. Furthermore, nowhere in this section is an explanation give for why an ice crystal, i.e.aspherical particle, should have a pulse width different than a droplet. I had fully expected that this section would give a detailed pulse shape analysis since I certainly could make an argument why the pulse SHAPE would look different for an aspherical particle, but I can’t find any physical reason why the width should differ.

Page 5767: Line 18. “…the PHDC in the panel (a) is poorly…”. There is no justification for saying that the PHD is poorly resolved and in fact, given that these are more or less mono-dispersed droplets, of 2 μm diameter, I would call this a remarkably well-defined PHD given the nature of the Gaussian beam. The main difference between the water droplet PHD and PWD and the ice crystal PHD and PWD is that the ice crystals are larger, having grown in ice saturated conditions. I can see nothing different between the two distributions that distinguish the ice from the water other than their size. This is exactly how most IN counters already differentiate the water from ice, so it seems that this proposed technique is not really unique.

How is the polarization feature being used? The title advertises polarization yet this is all about pulse width. Something got lost between the start of the paper and the end.

Page 5768: Line 9. “This mode of separation suggests an advantage of using the PWDC for the retrieval of ice fraction in the mixed-phase experiments. “ This was not convincing to this reviewer. It was neither shown theoretically why there should be a separation other than size, nor experimentally.

Page 5768: Section 4.2 The evaluation presented in this section is suspect due to the unconvincing nature of the pulse width approach. Figure 10 just illustrates that you can separate water from ice with the Welas just as easily as the PWD technique that is actually just separating by size (growth rate), as well.

References

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