

Interactive comment on “Probing ice-nucleation processes on the molecular level using second harmonic generation spectroscopy” by A. Abdelmonem et al.

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This is certainly a very interesting contribution that describes an exciting experimental technique that gives information on changes in water structure at a surface prior to freezing. I look forward to the more detailed study of the mica surfaces that the authors have promised!

There is one puzzling aspect of the results. I understand that the authors have identified the muscovite mica surface as the origin of the nucleation using a video camera. However, I am surprised that the basal plane of muscovite mica nucleates ice hetero-

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geneously in the immersion mode at a temperature as high as $-13\text{ }^{\circ}\text{C}$ (Figure 4). In a recent study of water droplets on mica, glass and silicon we found no experimentally significant difference in the freezing temperature of $50\text{ }\mu\text{m}$ diameter (approximately hemispherical) water droplets between the three surfaces (Campbell et al., 2015). For all three substrates the freezing temperature was $-35\text{ }^{\circ}\text{C}$, close to but slightly above the homogeneous freezing temperature of water. These results hence suggested that the mica basal plane is not a good ice nucleator in the immersion mode.

Moreover, in our experience ice nucleation on the basal plane of mica from vapour (deposition mode) almost always occurs in surface defects such as cracks or pockets under cleavage steps, as has also been found with many organic vapours nucleating on mica (Campbell et al., 2013).

As far as I am aware, most studies that show that mica is an efficient ice nucleator, whether in the deposition mode or in the immersion mode, such as the ones cited by the authors (Eastwood et al., 2008, and Glaccum and Prospero, 1980), have been conducted with ground mica or naturally occurring small flakes (e.g. in aerosols). It may be that other crystal planes exposed in the ground samples and flakes are the efficient nucleating agents, or that chemical effects arising from the leaching of ions via edges contribute. It should be noted, however, that in the study referred to above we did not find any noticeable enhancement of nucleation on scratching the basal plane of mica with diamond powder, which leads to the production of small flakes of mica. By contrast, such a procedure has proven very effective at enhancing nucleation from vapour with some organic substances (Holbrough et al. 2012). I think that it would be useful if the authors were to discuss in more detail the results of previous studies of the ice nucleating ability of muscovite mica.

I would also like some more technical detail on the contact angle measurements, particularly how the value of zero degrees for mica was obtained. What was the volume of liquid in the cell?

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