

Interactive comment on “Direct molecular level characterization of different heterogeneous freezing modes on mica” by Ahmed Abdelmonem et al.

Anonymous Referee #2

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This manuscript by Abdelmonem studies the deposition freezing of water on mica surfaces using SHG. The author claimed that two sub-deposition nucleation modes were identified but this claim was poorly supported by the experimental data. The manuscript also contains some incorrect statements about SHG. This reviewer does not recommend its publication in ACP.

(1) The author stated that “the SHG signal is originated from the nonresonant OH stretching vibrations at the interface”. (Line 8 on page 4) This statement does not make sense. A nonresonant signal is by definition not to be associated with a particular vibrational mode. (2) Mica is birefringent. As the laser beam goes through the mica, its polarization may not be linear any more. “The advantage of using SM polarization

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combination is its dependence on only one non-vanishing nonlinear susceptibility tensor element (line 33 on page 3)” is likely not what has happened in the experiment. (3) The index-matching gel of unknown chemical composition is a concern. While the gel helps to obtain the TIR condition, the gel/mica and gel/sapphire interfaces may produce SHG signal. The author may want to look into the freezing temperature of the gel too. (4) Figure 1 and 2 should be real-time plots similar to Figure 3. (5) Terms such as “liquid (film)”, “liquid (bulk)”, “transient ice”, “stable ice” used in Figure 2 should be experimentally defined. (6) The cooling rate dependence should be investigated. (7) One fundamental issue of SHG is that SHG intensity can be difficult to interpret. For example, why is there a $\sim 80\%$ SHG drop from the “air” to the “bulk liquid” in Figure 2? What does the SHG measure? (8) Some conclusions made in the manuscript are not well supported by the SHG data. For example, line 8 on page 4: “The coincidence of the SHG signals of the thin ice–film formed in DF2 and DF3 indicates identical structuring of water on the surface in two–step deposition freezing regardless of the onset temperature.” SHG simply cannot provide the structural information of water. The same SHG intensity does not necessarily mean the same water structure.

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