

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

**Anonymous Referee #1**

Received and published: 11 May 2017

This manuscript describes SHG measurements during water-mica interactions recorded at low temperature. The sparseness of the SHG data, the incorrect interpretation thereof, and the lack of complementary measurements lead this reviewer, unfortunately, to recommend rejection of the work. 1) The document is in a poor state of editing, with many grammatical errors that substantially distract from evaluating it. 2) The document contains false statements re: the origin of the SHG response. The system is probed off resonance, which means that all terms contributing to the response are purely real. Statements like "the SHG signal is originated from the the nonresonant OH stretching vibrations at the interface" are simply incorrect and reflect a fundamental misunderstanding of the signal generation process by the author. The

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signal is produced by all polarizable species within the SHG-active region. Unfortunately, the SHG active region is neither characterized nor defined in this work, making the signal interpretation at best appear as creative writing. 3) The interference that is briefly alluded to in the final paragraph is not quantified, even though the changes in the SHG responses shown in the three figures are produced by said interference, in addition to changes in surface potential that occur during the experiments. The author is encouraged to read and understand the recent work on nonlinear optical interference in thin-layer systems by Massari (J. Phys. Chem. Lett., 2016, 7 (1), pp 62–68) and on the chi(2) and chi(3) phase interference by Wang, Geiger, and Eisenthal (Nature Communications, 7, 13587, 2016). 4) The work requires additional information on ice layer thickness, on the uniformity of the ice layers across the 2 mm laser spot, and it requires verification whether the SHG signal depends quadratically on input power. The polarization states of the SHG responses during the various stages of the experiments should also be determined. 5) Connections of any results and/or discussion presented to the scientific motivation provided are not made except for two generic statements ("They provide novel molecular-level insight into different ice nucleation regimes..." and "Investigating the structuring of water molecules upon freezing next to solid surfaces is crucial to many scientific area...") which are broad and sweeping. In sum, this work is far too preliminary to be reconsidered. As such, this reviewer recommends rejection, with the hope that the author will write a new document that addresses the points made above in a new submission elsewhere.

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Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-322, 2017.

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