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Unraveling the microscopic pathway of homogeneous water crystallization at supercooled conditions from direct simulations FAUSTO MARTELLI, Department of Chemistry, Princeton University, JEREMY PALMER, Department of Chemical and Biomolecular Engineering, University of Houston, RAKESH SINGH, PABLO DEBENEDETTI, Department of Chemical and Biological Engineering, Princeton University, ROBERTO CAR, Department of Chemistry, Princeton University — By means of unbiased classical molecular dynamics simulations, we identify the microscopic pathways of spontaneous homogeneous crystallization in supercooled ST2 water. By introducing a new order parameter, we are able to monitor formation/disruption of locally ordered regions characterized by small ice clusters with intermediate range order. When two of these regions are close each other, they percolate and form a larger ordered region. The process is slow enough to allow for polymorphic selection in favor of cubic ice (Ic). The formation of an ice nucleus requires percolation of many small clusters so that the transformations at the interface of the nucleus do not involve its core, thus guaranteeing the stability of the nucleus. The growth of the crystalline nucleus is fast and involves direct transformation of interfacial liquid molecules as well as percolation of small Ic/Ih clusters. The growth is too fast to allow conversion of Ih into Ic sites, originating the formation of a stacking fault in the final crystal. We recognize Euclidean structures in the oxygen configuration of the second shell in Ic and Ih clusters. This new point of view allows us to explain the source of the ordered stacking fault geometry.

Fausto Martelli
Department of Chemistry, Princeton University