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Direct Calculation of the Rate of Homogeneous Ice Nucleation for a Molecular Model of Water AMIR HAJI-AKBARI, PABLO DEBENEDETTI, Department of Chemical and Biological Engineering, Princeton University — Ice formation is ubiquitous in nature, with important consequences in many systems and environments. However, its intrinsic kinetics and mechanism are difficult to discern with experiments. Molecular simulations of ice nucleation are also challenging due to sluggish structural relaxation and the large nucleation barriers, and direct calculations of homogeneous nucleation rates have only been achieved[1-2] for mW, a monoatomic coarse-grained model of water. For the more realistic molecular models, only indirect estimates have been obtained by assuming the validity of classical nucleation theory[3]. Here, we use a coarse-grained variant of a path sampling approach known as forward-flux sampling to perform the first direct calculation of the homogeneous nucleation rate for TIP4P/Ice, which is the most accurate water model for studying ice polymorphs. By using a novel topological order parameter, we are able to identify a freezing mechanism that involves a competition between cubic and hexagonal ice polymorphs[4]. In this competition, cubic ice wins as its growth leads to more compact crystallites[4]. [1] Li, et al., PCCP, 13, 19807 (2011) [2] Haji-Akbari, et al., PCCP, 16, 25916 (2014) [3] Sanz et al., JACS 135, 15008 (2013) [4] Haji-Akbari, Debenedetti, PNAS, 112, 10582 (2015)

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