Supporting Information to "Homogeneous ice nucleation at moderate supercooling from molecular simulation"

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Equilibration of the initial configuration

The preparation protocol used for all cluster sizes is the following. After having inserted the ice cluster in the supercooled liquid, we remove the liquid molecules overlapping with the solid ones. Next, we equilibrate the system for about 0.2 ns at 200 K. To make sure that the chosen 0.2 ns is a proper equilibration time, long enough to allow for annealing mismatches at the interface, we run a simulation starting from the initial configuration at time zero. In Fig. S1, we represent the cluster size for the Large system of the TIP4P/2005 model as a function of time. The figure shows that,

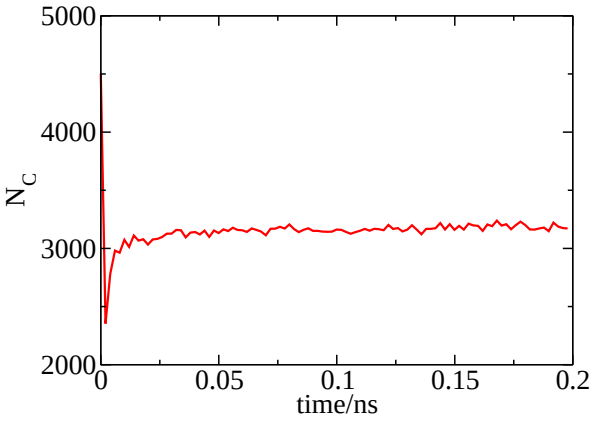

Figure S1: Equilibration for the L system of the TIP4P/2005 model. After an initial drop of the cluster size due to equilibration of the interphase, the cluster size changes very slowly with time.

even though the dynamics is very slow at such low temperatures, the chosen equilibration time is long enough. This result is independent on the chosen cluster size or water model potential.

Choice of the order parameter to distinguish between liquid/solid particles

The use of an alternative order parameter to identify solid-like particles ($\bar{q}_3$) does not affect the observed response of the cluster to temperature. This is shown in Fig. S2, where the number of particles in the cluster is monitored with two different order parameters for three different temperatures. Both order parameters allow to conclude that the inserted cluster is critical between 235 and 240 K.

Obviously, the number of particles that belong to the cluster does depend on the order parameter. The order parameter we use in this work should at least work well for the inner particles given that, according to Fig. 1 of the main text, it is able to discriminate between bulk liquid and bulk solid particles. The main ambiguity in the number of particles belonging to the cluster comes from those particles that lie in the interface. In view of Fig. 3 of the main text, it seems that our order parameter is doing reasonably well in identifying such particles either. Nonetheless, we have considered an error as large as 60% in the identification of the *interfacial particles* to estimate the error of the nucleation rate. In this way the unavoidable ambiguity in the determination of the cluster size is reflected in the error bar of J.

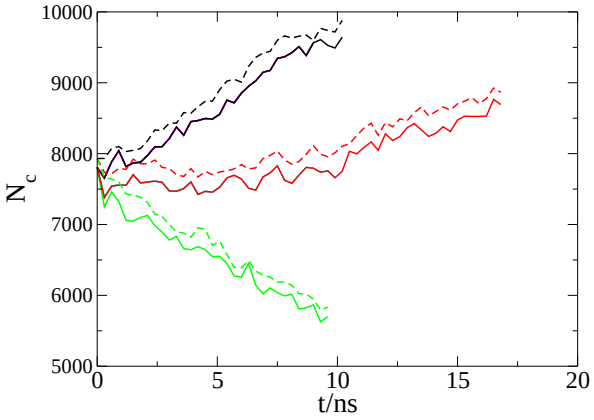

Figure S2: Number of particles in the cluster versus time for system size H, TIP4P/2005 model. Black curves correspond to 230 K, red ones to 235 K and green to 240 K. Solid lines correspond to the analysis made with the order parameter described in the main text. Dashed lines correspond to the use of an alternative order parameter. With such order parameter particles are considered as neighbors if their oxygen atoms are closer than 3 Å and are labelled as solid-like whenever their $\bar{q}_3$ is larger than 0.28.

Cluster size and potential energy versus time for all system sizes and model potentials studied

In order to determine the temperature at which the cluster was critical, we evaluated the highest temperature at which the cluster grows and the lowest temperature at which it melts. In Fig. 4 of the main text we represented the number of molecules in the ice cluster versus time for system H simulated

with TIP4P/2005 potential. In what follows, we present the results of the cluster size versus time for all sizes (B,L and H) for both the TIP4P/2005 (Fig.S3) and TIP4P/Ice water models (Fig.S4). An analogous result could have been obtained by monitoring the potential energy of the system as a function of time (see the right panels at Figs. S3 and S4).

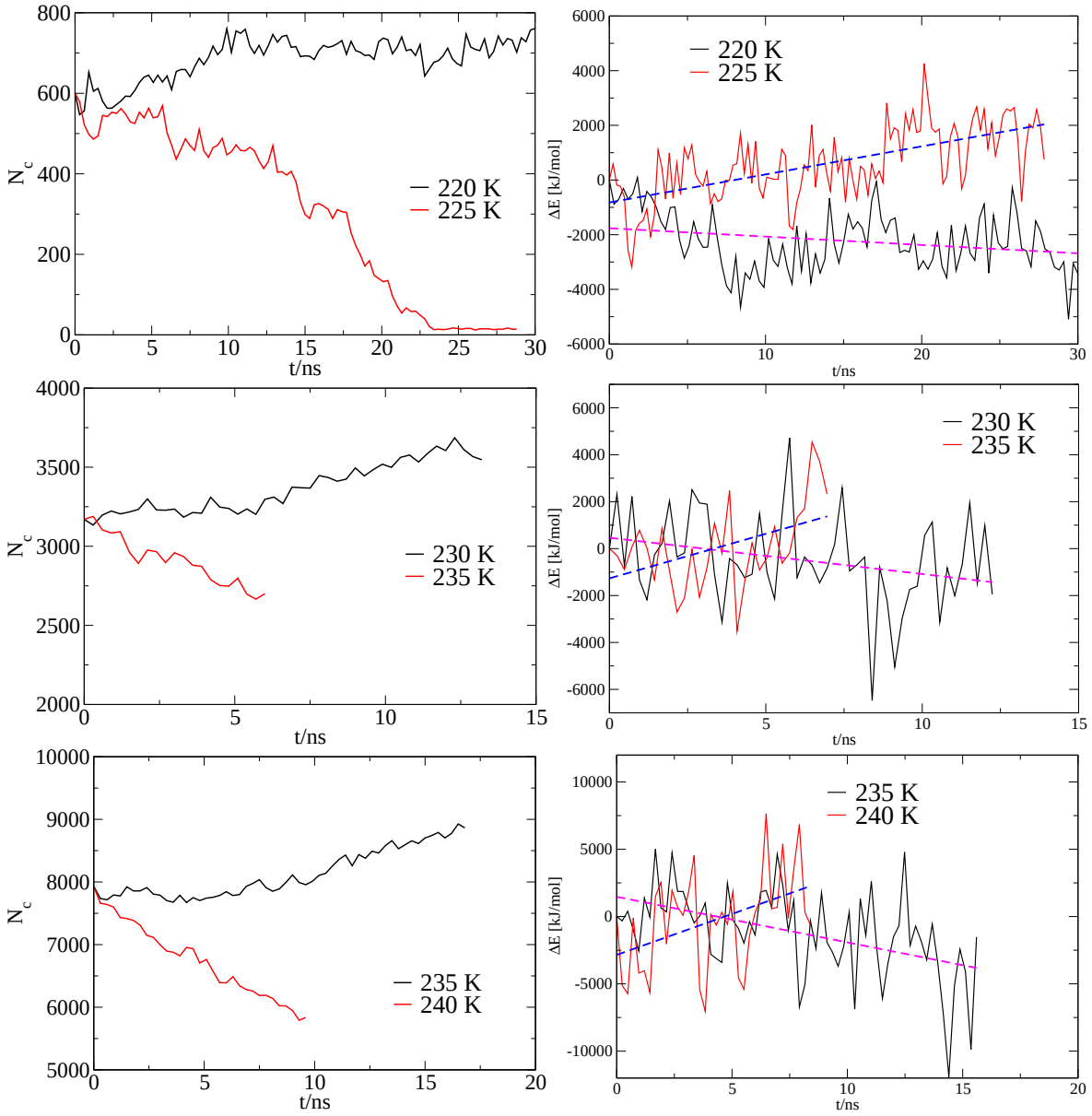

Figure S3: Left-hand panels: Number of molecules in the ice cluster versus time for system B (top), L (middle) and H (bottom) simulated with the TIP4P/2005 potential. Right-hand panels: energy difference (between the energy at time t and the one at time zero) versus time. Results are shown for different temperatures as indicated in the legend.

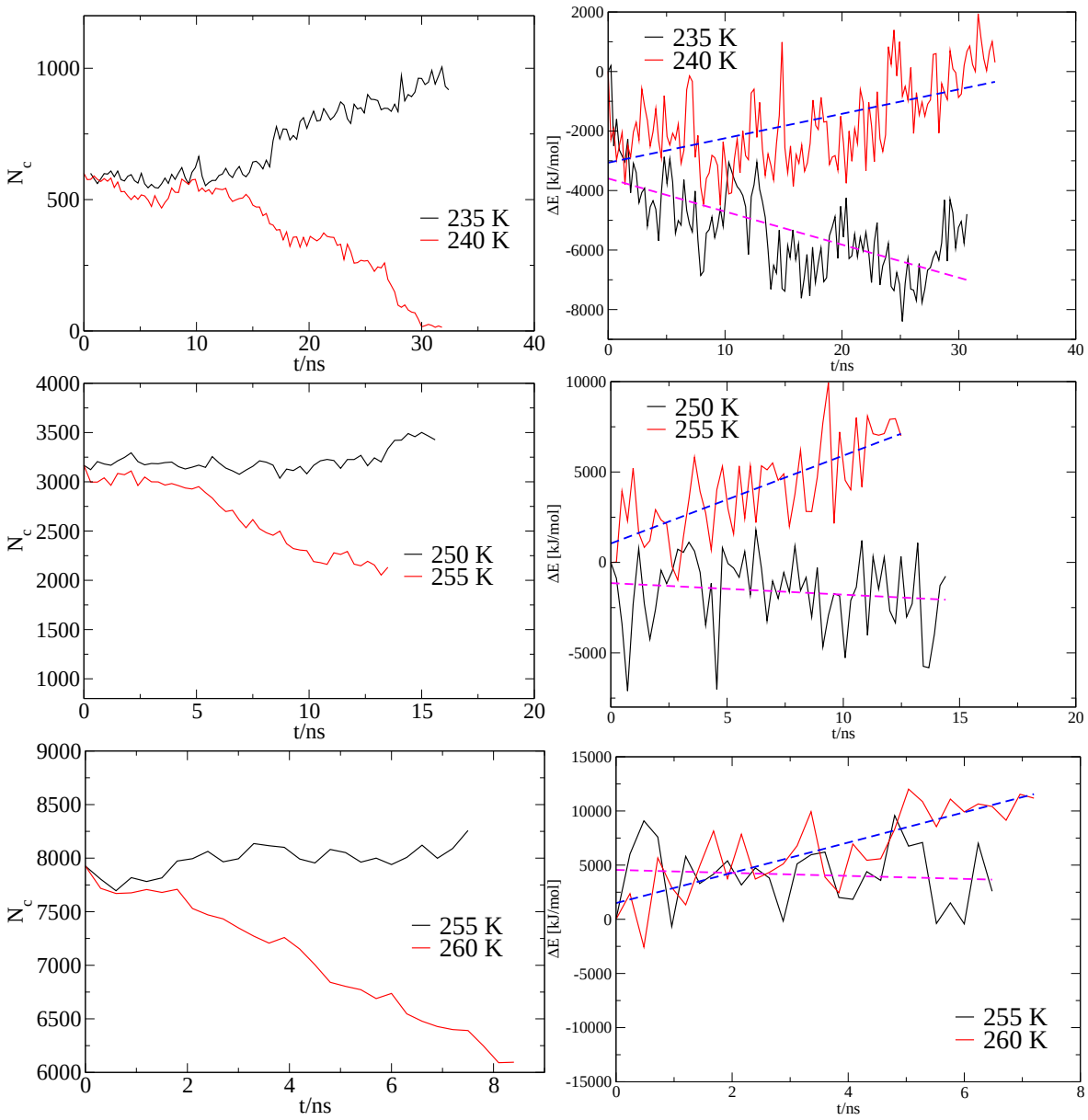

Figure S4: Same as Fig. S3 but for TIP4P/Ice instead of TIP4P/2005.

The energy is much less sensitive to changes in the cluster size than the order parameter. This is due to the fact that the number of molecules in the cluster is a small fraction of the total number of molecules. Nonetheless, by making a linear fit to the time evolution of the energy we obtain in all cases consistent results with the analysis based in the order parameter: the cluster grows when the slope is negative and shrinks when it is positive.