Ice growth rate: Temperature dependence and effect of heat dissipation

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ABSTRACT

The transformation of liquid water into solid ice is arguably the most important phase transition on Earth. A key aspect of such transformation is the speed with which ice grows once it is nucleated. There are contradictory experimental results as to whether the ice growth rate shows a maximum on cooling. Previous simulation results point to the existence of such a maximum. However, simulations were performed at constant temperature with the aid of a thermostat that dissipates the heat released at the ice-water interface unrealistically fast. Here, we perform simulations of ice growth without any thermostat. Large systems are required to perform these simulations at constant overall thermodynamic conditions (pressure and temperature). We obtain the same growth rate as in previous thermostatted simulations. This implies that the dynamics of ice growth is not affected by heat dissipation. Our results strongly support the experiments predicting the existence of a maximum in the ice growth rate. By using the Wilson-Frenkel kinetic theory, we argue that such maximum is due to a competition between an increasing crystallization thermodynamic driving force and a decreasing molecular mobility on cooling.

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I. INTRODUCTION

The transformation of supercooled water into ice is one of the most important phase transitions on Earth. It involves two stages. Initially, a process known as nucleation occurs, where a solid ice nucleus must appear after overcoming a free energy barrier. Nucleation can be heterogeneous in the presence of impurities[1–5] or homogeneous[6–8] in the case of pure water. The emergence of a critical ice nucleus enables other molecules to join. That is the second stage known as crystal growth, and it is the framework of this article.

Understanding and, hence, learning to control the growth of ice are important for various and diverse disciplines such as climate science (precipitation and albedo),[9] aviation technology,[10] cryopreservation,[11,12] food industry,[13] or materials science.[14]

Despite the importance of water and the ubiquitous character of the water-to-ice transition, there are still large discrepancies and uncertainties in experimental measurements of the ice growth rate.[15–19] There are even qualitative differences in

the temperature dependence of the growth rate, whereas some experiments support the existence of a maximum[19] and some others deny it.[17]

Fortunately, experimental research can be complemented with computer simulations. Nowadays, there are several water models that accurately describe the behavior of real water.[20–22] Simulation estimates of the ice growth rate using different water models[23–28] point to the existence of a maximum in the ice growth rate.

However, there is one remaining issue that questions the validity of these simulations: they were carried out at constant temperature with the aid of a thermostat that dissipates the freezing heat released at the interface as ice grows within a small relaxation time (typically a few picoseconds). One may wonder whether an artificially quick heat dissipation due to the use of a thermostat may affect the dynamics of ice growth.[18,27]

In this paper, we compare simulations of ice growth previously performed in the NpT ensemble (with thermostat) with new ones performed in this work in the NVE ensemble (with no thermostat). Ice growth simulations in the NVE ensemble are more akin

to experiments because the heat released by freezing is not removed by a thermostat. Using the NVE ensemble poses a computational challenge, though, given that large systems are required to avoid temperature or pressure drifts during the course of the simulation.

Within the accuracy of our simulations, we find that the use of a thermostat does not alter the rate of ice growth. This implies that latent heat dissipation is faster than ice growth. Thus, our work confirms the existence of a maximum in the ice growth rate predicted in previous simulations and serves as a guide to solve the discrepancies between different experimental measurements. We also check that the Wilson-Frenkel theory[29–32] quite accurately predicts the growth rate estimated in our simulations. Therefore, one can understand the maximum in the growth rate as a result of the competition between thermodynamics (chemical potential difference between both phases) and kinetics (diffusion coefficient).

II. METHODS AND SIMULATION DETAILS

Molecular dynamics simulations were performed using the GROMACS package,[33] while water was simulated with the TIP4P/Ice model.[34] Such a model accurately reproduces water properties especially when solid phases are present and its melting point is 270 K,[26,27,35] very close to the experimental value of 273.15 K.[36] The computational work had two main stages. The aim of the first one was to prepare well equilibrated configurations. A small block of ice containing $N_{ice}^{i} \approx 1000$ water molecules was set in contact with a large bath of $N_{bath}^{i} \approx 20\,760$ water molecules. The total number of molecules involved in the simulation was always $N = 21\,760$. During this stage, simulations were done in the NpT ensemble in order to set temperature and pressure at the values of interest. These are pressure always $P \sim 1$ bar and temperatures $T = 265, 257, 251,$ and 243 K corresponding to supercoolings $\Delta T = 5, 13, 19,$ and 27 K. The second stage consisted in simulations in the NVE ensemble of the configurations prepared in the first stage. After the preparation, N_{ice}^{i} as well as N_{bath}^{i} could have slightly changed so that these initial values during the production stage depend on the run. The plane that the ice block exposed to the liquid water was the secondary prismatic $\{1\bar{2}10\}$. A representation of the system that was created with VMD[37] is shown in Fig. 1.

In both the preparation and production stages, the LJ term of the potential and the real part of the electrostatic potential was truncated at 12.5 Å; standard tail corrections were added beyond

that distance both to energy and pressure. We used PME[38] to deal with the long range electrostatic corrections with 0.1 nm Fourier spacing and a 4th order interpolation polynomial. In order to constraint the geometry of the molecule, the LINCS algorithm[39] was used.

During the NpT simulation stage, a velocity-rescale thermostat[40] and an anisotropic Parrinello-Rahman barostat[41] were used with a relaxation time of 0.5 ps in both cases. The time step was set to 2 fs and runs lasted until the systems were equilibrated (around 2 ns). In the NVE simulations, it was crucial to use a short integration time, 0.2 fs in our case, to maintain the energy constant along the trajectories. Runs lasted until the size of the ice block was almost twice the initial one for a given trajectory. The time needed for this was 13 ns for the fastest case (257 K) and 22 ns for the slowest case (242 K). Both the short integration time step and the large system size required make these simulations a computational challenge. We needed a 24-core machine for about a month in order to get a single estimate of the growth rate. We obtained the growth rate for 4 different state points. Two independent estimates were made for each state point to improve statistics.

Concerning the analysis of the simulations, once the ice block had grown sufficiently, we obtained the growth rate u as

$$u = \frac{\Delta L_{ice}}{2\tau}, \tag{1}$$

where ΔL_{ice} is the distance grown by ice and τ is the simulation time. The factor of 2 comes from the fact that there are two interfaces growing under periodic boundary conditions. ΔL_{ice} can be estimated as

$$\Delta L_{ice} = \frac{\Delta N_c}{\rho_{ice} A}, \tag{2}$$

where ΔN_c is the number of molecules that incorporate into the ice phase during the simulation, A is the area of the section, and ρ_{ice} is the ice number density at (T, P).

To compute the number of molecules in the ice phase, N_c, we employed the widely used $\bar{q}_6$ order parameter[42] with a cutoff of 3.5 Å to label icelike and liquidlike molecules in the system (see Refs. 43–45 for details on the implementation of the $\bar{q}_6$ order parameter for water). An example of the evolution of N_c during the course of one of our simulations is given in Fig. 2. The slope of the curve is proportional to the growth rate.

(a)

(b)
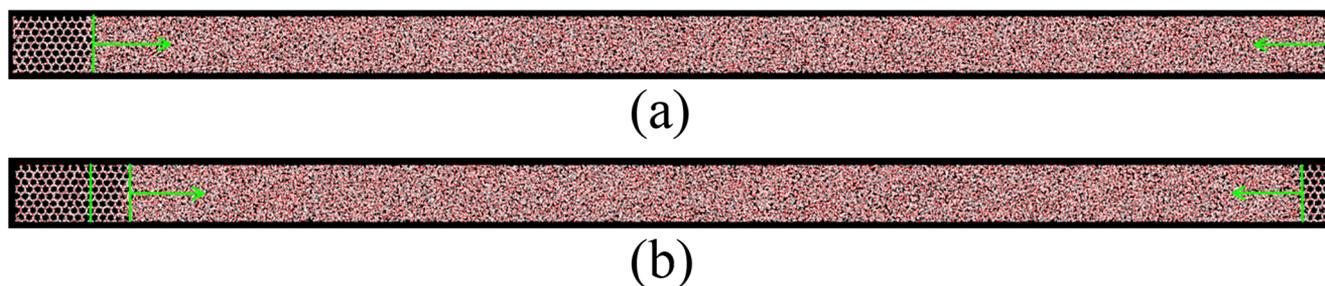
FIG. 1. (a) Initial configuration of a typical simulation for computing the growth rate, being the thermodynamic conditions 257 K and 1 bar. (b) The same system after 13 ns, showing how both the left and right interfaces have moved toward the liquid. The plane exposed to the liquid is the secondary prismatic $\{1\bar{2}10\}$.
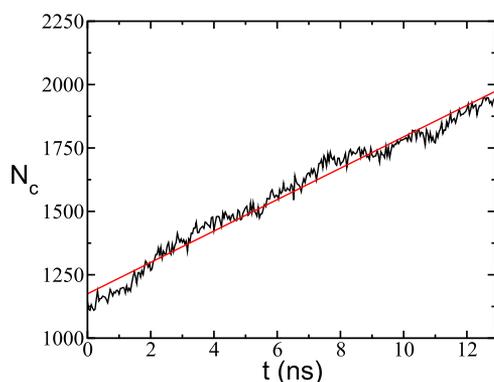
FIG. 2. Number of particles in the biggest cluster of icelike molecules, N_c, as a function of time for $T = 257$ K.

III. RESULTS

A. Validity of the NVE calculations

As explained in the Introduction, we use the NVE ensemble to simulate the dissipation of freezing heat during ice growth more realistically than when a thermostat is employed. Working in the NVE ensemble is tricky, though. To avoid significant energy, pressure, or temperature drifts, and hence be able to compare our results with experiments or thermostated simulations, the molecular dynamics time step must be short and the system must be large (see Sec. II). In particular, the number of water molecules that incorporates into the ice phase during the simulation must be small as compared to the total number of liquid molecules (see Fig. 1 for a snapshot). In this way, neither the heat released by freezing will change much the overall temperature nor the expansion due to freezing will cause a large pressure raise.

To assess the validity of our NVE simulations, we follow the evolution of the total energy, temperature, and pressure. In Fig. 3, we show these variables for one trajectory initially at 257 K and 1 bar. As Fig. 3(a) shows, the total energy in the system is well conserved. Temperature is shown in Fig. 3(b) and increases about 2 K along the course of the 13 ns simulation due to the freezing process. The growth rate determined in the simulations is assigned to the average temperature of the run rather than to the initial one. Consequently, we have a temperature uncertainty of about ±1 K in each determination of the growth rate. Note that if the runs were not interrupted and crystallization continued, the temperature would keep raising and, at some point, the growth rate would change according to the new thermodynamic conditions. Thus, the analysis must be restricted to time intervals where the growth rate is steady. In Fig. 3(c), we show the variation of pressure. Since the simulation volume is fixed and ice occupies more space than liquid water, pressure increases as the ice front moves forward. Another contribution to this effect could be brought by an increase in temperature. Anyway, the pressure raise is small and comparable to the fast, local pressure fluctuations. With this analysis, we conclude that our simulation set up enables keeping pressure and temperature roughly constant during the time required to measure the growth rate.
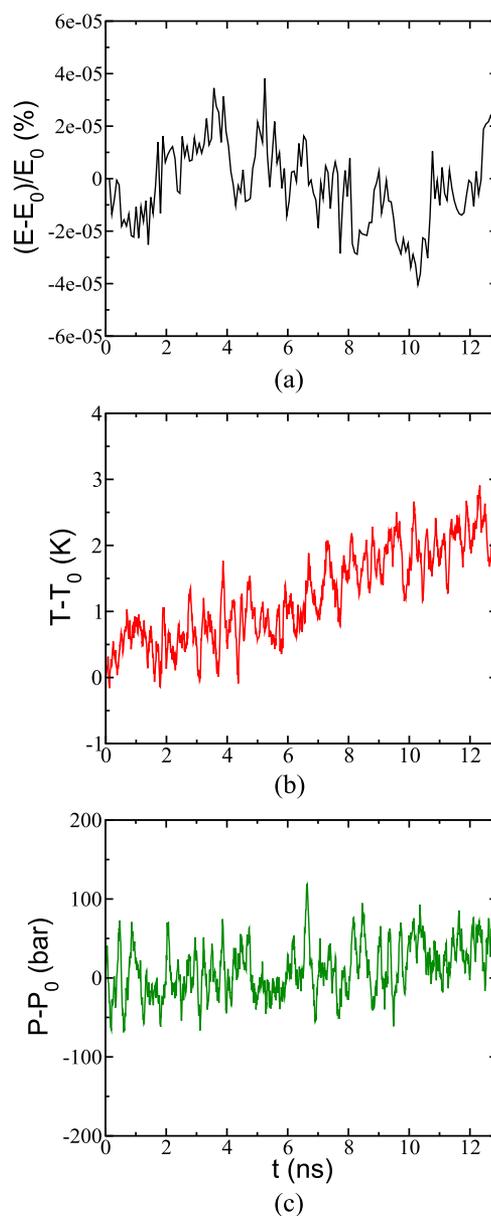
FIG. 3. Time evolution of the relative change (in %) of the total energy (a), the temperature (b), and the pressure (c) for a system prepared at 257 K and 1 bar. All the panels share the same horizontal axis.

B. NVE vs NpT

Having made sure that pressure and temperature are kept fairly constant along our NVE simulations, we can compare our results for the growth rate with those obtained by means of NpT simulations. Such comparison is established in Fig. 4(a). Red symbols are the NVE results of this work, and black and blue ones correspond to NpT simulations from Refs. 18, 26, and 27. In all cases, the TIP4P/Ice water model was used and the secondary
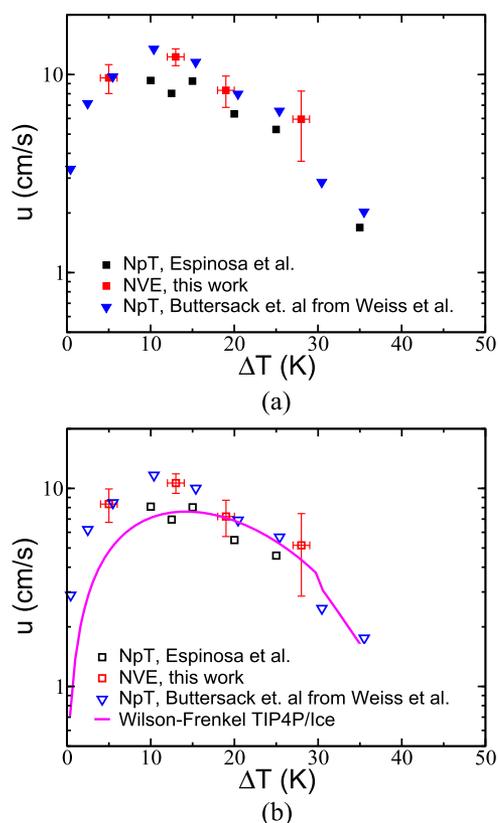
FIG. 4. (a) Growth rate of ice in the secondary prismatic direction as a function of supercooling, using the TIP4P/Ice model, both in the NVE (this work) and NpT ensembles. Black squares have been taken from Ref. 26. Blue triangles are a private communication from the authors of Ref. 27 to those of Ref. 18. Error bars represent standard error. In (b), simulation data presented in (a) are multiplied by a factor of 13/15 in order to obtain an estimate of the growth rate averaged over all crystalline directions, as suggested in Ref. 26. The magenta line corresponds to the Wilson-Frenkel theory using the TIP4P/Ice parameters (see main text).

prismatic ice plane was exposed to the liquid. As shown in Fig. 4, the results of both simulation ensembles are in very good agreement. The main conclusion that can be drawn from this successful comparison, which is the main result of this work, is that the freezing heat dissipation does not affect the dynamics of ice growth. This means that the time scale associated with the dissipation of the heat released in the interfacial region when ice grows is much shorter than that related to ice growth. In fact, as shown in Fig. 5, there are no significant temperature gradients along the direction perpendicular to the interface in the time scale of ice growth.

C. Maximum in the growth rate

In Fig. 4(a), we show that the growth rate has a maximum for the TIP4P/Ice model. The same trend has been observed by Rozmanov et al. using the TIP4P/2005 model.[24,25] In this work, we show that such behavior is robust and independent of the use of a thermostat in the simulations.
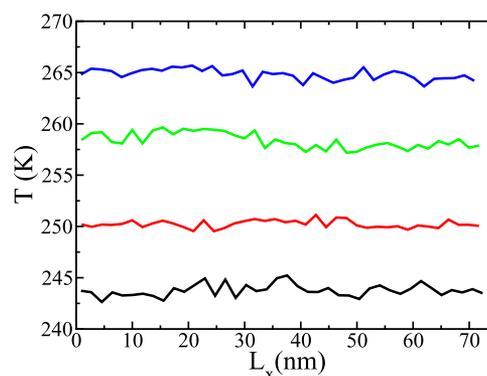
FIG. 5. Temperature profile along the direction perpendicular to the interface for all systems simulated averaged over the last half of the simulation.

Such maximum can be understood as a consequence of the competition between opposing thermodynamic and kinetic effects. On the one hand, upon cooling, the solid becomes increasingly more stable than the liquid, thus raising the thermodynamic driving force for ice growth. On the other hand, lowering the temperature slows down diffusion[46] and molecules take longer to incorporate into the growing ice phase. These competing factors give rise to the Wilson-Frenkel theory for the growth rate,[29–32]

$$u(T) = \frac{D(T)}{a}\left[1 - \exp\left(-\frac{|\Delta\mu(T)|}{k_B T}\right)\right], \tag{3}$$

where $D(T)$ is the diffusion coefficient, a is a characteristic length—of the order of the molecular diameter—$\Delta\mu$ is the chemical potential difference between the solid and the liquid, and k_B is the Boltzmann constant. We can test the theory by using Eq. (3) with the parameters of the TIP4P/Ice model. $D(T)$ and $\Delta\mu(T)$ are given in Ref. 26 and we take $a = 3$ Å, about one molecular diameter. The theory gives the magenta line in Fig. 4(b), which describes quite well the trend of the TIP4P/Ice simulation data, particularly so beyond the maximum. (The kink in the line of the Wilson-Frenkel theory is due to the fact that in Ref. 26, two different fits to the diffusion coefficient were used, one at moderate supercooling and another at large supercooling.) We stress that the magenta line is not a fit, but the theory combined with simulation parameters. Therefore, the view that the maximum in the growth rate is due to a competition between an increasing thermodynamic driving force and a decreasing mobility seems to be correct.

D. Experiments

In Fig. 6, we plot the results for the ice growth rate from different experimental groups. As mentioned in the Introduction, there are strong discrepancies. Seminal work by Pruppacher et al.[16] was restricted to low supercooling, where there is no sign of a maximum in the growth rate (green triangles). Pruppacher's work is in good agreement with that by Buttersack et al.[18] (turquoise triangles). The work by Buttersack et al.,[18] that extends to deeper supercooling, seems to be consistent with a maximum, although their data are quite scattered and no definite conclusion can be drawn in this
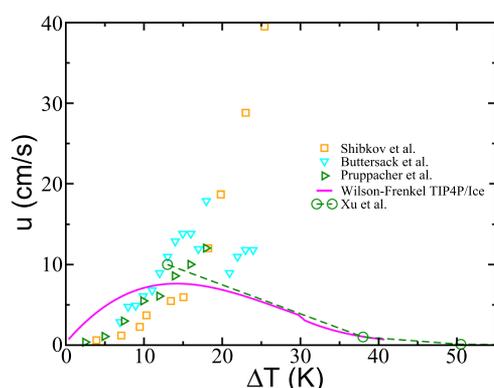
FIG. 6. Rate of ice growth u as a function of supercooling. Symbols correspond to different experiments: laser-pulsed nanofilms by Xu et al.[19] (empty green circles connected by dashed green lines as a guide to the eye), thin layers by Shibkov et al.[17] (empty orange squares), droplets by Buttersack et al.[18] (cyan inverted triangles), and Pruppacher et al.[16] (dark green triangles). The magenta line corresponds to the Wilson-Frenkel theory applied to the TIP4P/Ice water model as explained in the main text. This line describes quite well the simulation results for u [see Fig. 4(b)].

respect. Shibkov et al.,[17] however, did not find a maximum but a sharp monotonic increase of the growth rate with the supercooling (orange squares). This result is in stark contrast with the work by Xu et al.[19] (green circles), who performed experiments up to very deep supercooling with water nanofilms. They see a monotonic decrease of the growth rate with the supercooling, a tendency consistent with the maximum.

In Fig. 6, we include the Wilson-Frenkel curve previously presented in Fig. 4(b) that describes pretty well the trend for the TIP4P/Ice simulation data. Such line is in very good agreement with the experiments by Xu et al. This agreement clearly supports the experiments that claim the existence of a maximum in the growth rate.

Before our work, both simulations and experiments claiming the existence of such a maximum were questioned for the way heat dissipation was dealt with. On the one hand, in simulations, the use of a thermostat almost instantly dissipates the heat released at the interfacial region while ice grows. On the other hand, the experiments by Xu et al. are performed with ultrathin water films (nanometer thick), which would enable a faster heat dissipation than in experiments with large samples. Therefore, both the use of thermostats or of thin samples questioned the validity of the experimental and simulation works that support the existence of the maximum in the growth rate. However, the good agreement between NVE and NpT simulations shown in Fig. 4 proves that heat dissipation does not affect the ice growth dynamics. Therefore, our work is a strong support for the existence of a maximum in the ice growth rate. Our simulations with the TIP4P/Ice model predict the maximum to occur at 14 K below melting and at a growth rate of ~10 cm/s.

A comment is due on the comparison between simulations and ice growth experiments in large samples.[16,17]

There, the mechanism of growth is dendritic and several fronts simultaneously grow. In this situation, only ~one third of the molecules initially engulfed in the dendritic branch is in the crystalline state.[18] The rest of the water molecules inside the dendritic ice network take longer time to crystallize. In our simulations, we estimate the speed of growth of a single front growing in detriment of the liquid. Note that dendritic developments cannot happen even when simulating the secondary prismatic plane due to the limited size of the simulation box. In any case, our results disagree with those of Shibkov et al.,[17] since even if our system related to the slowest front in their dendritic front, it would not reach their reported growth rate. For instance, at 25 K of supercooling, they found a freezing velocity of 40 cm/s, while in our calculations we find around 5 cm/s.

IV. SUMMARY AND CONCLUSIONS

We investigate the effect of heat dissipation in the dynamics of ice growth. For that purpose, we perform NVE simulations of a growing ice front in contact with supercooled water and compare the results with previous estimates in the NpT ensemble, where freezing heat dissipation is artificially fast due to the effect of a thermostat. First, we validate our NVE simulations that must be performed with large systems and short time steps in order to avoid temperature and pressure drifts. Our NVE results are fully consistent with those previously obtained in the NpT ensemble. This agreement leads us to conclude that ice growth is not affected by the dissipation of freezing heat from the interfacial region. This statement implies that the heat dissipation is much faster than ice growth. Our simulations support some experiments that show the existence of a maximum in the growth rate as a function of the supercooling and disagree with some others that predict a monotonic increase of the growth rate on cooling. We successfully apply the Wilson-Frenkel theory for crystal growth rates to the employed water model. The good performance of the theory suggests that the maximum in the growth rate is due to a competition between an increasing thermodynamic driving force for crystallization and a decreasing mobility of the molecules on cooling.

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