Supplementary Information for

Nanoscale insights into vibration-induced heterogeneous ice nucleation

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S1 New LAMMPS fix and Conservation of energy



Figure S1: (a) Temperature of water as a function of time; (b) Variation of different components of energy associated with the system over time; (c) Comparison between the total energy change ΔE_{total} in the system and the cumulative work done W_d by the vibrating surface. (ΔPE_{water} and ΔKE_{water} represent the changes in potential energy and kinetic energy, respectively. The subscripts *water*, *low*, and *up* represent the confined water between the surfaces, the lower surface, and the upper surface, respectively.)

As mentioned in the paper, the current study assumes constant temperature throughout the simulation process, as the vibrating surface mimics a portion of a larger bulk material which would act as a heat sink in a realistic application. During surface vibration, the work performed by the surface dissipates and accumulates as heat in the system, which must be removed to maintain a constant temperature. To achieve this, a thermostat is applied to the surface rather than the water. This approach minimizes the influence of the thermostat on nucleation behavior. In realistic metal-water interfaces, the surface typically possesses much higher thermal conductivity than the liquid, allowing it to efficiently extract heat from the system. However, the default LAMMPS class (fix move) for surface vibration constrains surface atoms to move strictly according to a sine curve, as described in Equation 1 of the main paper, and preventing the necessary thermal velocity to be superimposed on the vertical sinusoidal motion. This limitation affects the thermal conductivity at the water-surface interface as well as the surface itself. To address this issue, we developed a new LAMMPS class, fix movenve, which enables a group of atoms to exhibit center-of-mass sinusoidal motion while simultaneously maintaining thermal velocity during time integration. This modification ensures that a surface thermostat can effectively regulate the surface temperature.

To validate our newly developed LAMMPS fix, we track the temperature and energy changes during the simulation. Figure S1 illustrates the temporal variation of temperature and energy for a representative case (T = 220 K, f = 100 MHz, A = 12 Å). As shown in Figure S1(a), the mean temperature of the water remains stable around 220 K throughout the 100 ns simulation with surface vibrations, with only minor fluctuations around the mean. Figures S1(b) and (c) assess energy conservation by tracking the evolution of different energy components associated with the system and comparing the total to the cumulative work done by the vibrating surface. According to the first law of thermodynamics, the total energy change, ΔE_{total} , of the system should equal the cumulative work done, W_d , by the vibrating lower surface ($\Delta E_{\text{total}} = W_d$). This relationship is confirmed in Figure S1(c).

Note that ΔE_{total} is calculated by

$$\Delta E_{total} = \Delta P E_{water} + \Delta K E_{water} + \Delta P E_{low} + \Delta K E_{low} + \Delta P E_{up} + \Delta K E_{up} + \Delta Q_{low} + \Delta Q_{up},$$
(1)

where ΔPE and ΔKE are the changes in potential energy and kinetic energy, respectively, while ΔQ represents the heat removed by the thermostats at the surfaces. The subscripts *water*, *low*, and *up* refer to the confined water between the surfaces, the lower surface, and the upper surface, respectively. The work done W_d is calculated by

$$W_d = \sum_{t=1}^{N_t} \frac{F_{loc,t} + F_{loc,t-1}}{2} \cdot \Delta d,$$
 (2)

where N_t is the total number of timesteps, $F_{\text{loc},t}$ and $F_{\text{loc},t-1}$ are the local forces exerted on the lower surface by the water at timesteps t and t-1, respectively, and Δd is the displacement between timesteps t and t-1. The excellent agreement of ΔE_{total} and W_d in Fig S1(c) validates the use of *fix movenve* in this work. Note that the small deviations in temperature observed in Figure S1(a) do not influence the nucleation process, as elaborated in Section S7.

S2 Setting simulation temperature

Ice nucleation is a rare event, and classical MD simulations typically requires extended runtimes to capture it. A long runtime allows molecules to explore a larger portion of



Figure S2: (top) Temperature ramp on the water from 290 K to 200 K and (bottom) resulting potential energy variation, for all three realisations. The nucleation temperature can be determined by tracking when the potential energy drops.

phase space and overcome the energy barrier to nucleation. However, these runtimes can exceed available computational resources. A common approach to address this challenge is brute-force simulation, where significant supercooling is applied to lower the nucleation energy barrier, enabling nucleation within MD timescales. However, it is crucial to ensure that the supercooling does not approach or exceed the spinodal temperature (the theoretical limit for water remaining in the liquid phase). If the temperature is too low, nucleation may occur spontaneously at the start of the simulation rather than as a consequence of surface vibration.

To identify a suitable temperature for brute-force ice nucleation, we conducted a separate set of preliminary simulations. These simulations used the same system described in the paper but kept the surfaces static (no vibrations were applied). A Nosé–Hoover thermostat was applied to the confined water layer (238 Å thick) to cool the system from 290 K to 200 K at a rate of $k_c = 1$ K/ns. Three independent realizations were performed, each beginning with different equilibrated configurations. The results show that ice nucleation is accompanied by a sudden jump in the system's potential energy, as illustrated in Figure S2. We identified these inflection points in the potential energy curve for each realization and averaged the corresponding nucleation induction times. The nucleation temperature, T_{ind} , was calculated by $T_{ind} = 290 - k_c \cdot t_{ind} = 209.5$ K (obtained from Figure S2(top)), where $t_{ind} = 80.5$ ns is the averaged nucleation induction time. For the vibration simulations, we set the system temperature to be 10.5 K higher than T_{ind} , to T =220 K. This elevated temperature increases the nucleation energy barrier, reducing the likelihood of nucleation in the absence of external intervention (vibration in the current case). This setup allows us to study how vibration influences nucleation on the static surface under controlled conditions.



S3 Calculating nucleation rate

Figure S3: Calculation of nucleation rates based on survival probability for three representative cases (100 MHz, 16 Å; 300 MHz, 16 Å; and 500 MHz, 16 Å). (a) Variation of potential energy with time. Induction time t_{ind} of ice nucleation is estimated by fitting the red line based on Equation 3. The insets show snapshot of the simulation domain before, during, and after ice nucleation. (b) Variation of P(t) with time. The continuous lines represents the fitting of Equation 5 to estimate nucleation rate J.

To ensure that the conclusions drawn from the regime map in the paper were accurate, we performed several additional validation checks, such as the accurate estimation of nucleation rates. We calculated nucleation rates for three representative cases from different regimes: complete nucleation (100 MHz, 16 Å), partial nucleation (300 MHz, 16 Å), and no nucleation (500 MHz, 16 Å), as indicated by star-shaped symbols in the regime map shown in the inset of Figure S3(b). The nucleation rate is determined based on the survival probability P(t) from 15 simulations under specific icing conditions, following these steps:

A) The potential energy of the water molecules is tracked during the simulations (Figure S3(a)). A sudden drop in energy, corresponding to the phase change, is racked and the corresponding time t is recorded. Equation 3 is fitted to the data to obtain the induction time (t_{ind}) , minimizing statistical and thermal noise:

$$E(t) = a + \frac{b}{1 + \exp(c(t - t_{\text{ind}}))}.$$
(3)

Here, a, b, and c are fitting constants.

B) The induction time (t_{ind}) is estimated from 15 simulations under identical conditions. This provides a distribution of t_{ind} for each specific icing condition.

C) The distribution of t_{ind} is then used to calculate the survival probability using Equation 4. The time versus survival probability relationship is shown in Figure S3(b):

$$P(t) = 1 - \frac{1}{N_{\rm sim}} \sum_{i=1}^{N_{\rm sim}} \Theta(t - t_{\rm ind}^i).$$
(4)

Here, Θ is the Heaviside step function, and $N_{\rm sim}$ is the number of simulations.

D) The time versus survival probability curve is fitted to a Poisson process model (Equation 5) to calculate the nucleation rate (J):

$$P(t) = \exp(-(Jt)^{\gamma}). \tag{5}$$

Further details on the nucleation rate calculation are available in Cox et al¹. Note that for cases where no ice nucleated during the 100 ns simulation, the nucleation time was treated as *censored data*, meaning it was excluded from the calculation of P(t). As expected from the regime map, significant differences in nucleation rates were observed among the three cases. The case in the complete nucleation regime exhibited the highest nucleation rate of 0.01585 ns⁻¹, while the case in the no nucleation regime showed the lowest rate of 0.00097 ns⁻¹. These nucleation rates are consistent with the regime map, highlighting the varying degrees of enhancement induced by surface vibrations. Note that no nucleation is observed in the absence of surface vibrations.

S4 Relaxation times of water model underlying f_c/T_c

A critical period of $T_c = 6.67$ ns (or conversely a transition frequency of $f_c \approx 150$ MHz) was observed in our simulations, below which ice nucleation did not occur. Our hypothesis was that once the surface vibration is rapid enough, i.e. once $f > f_c$, the water molecules no longer have sufficient time to rearrange to form ice, so T - c likely has a molecular origin. To validate this, we conducted additional molecular dynamics analyses to explicitly quantify the relaxation time of supercooled water molecules after a (sudden) transient acoustic perturbation. The simulation setup for these additional analyses is depicted in Figure S4(a). This setup was identical to the one employed in the main manuscript in terms of domain dimensions and components, with one key difference: the lower substrate, instead of undergoing continuous harmonic vibrations, was instantaneously displaced downward by a distance equal to the vibration amplitude used in the main study. This instantaneous displacement simulated a rapid acoustic perturbation. The displacement was executed with a pulling speed of 500 Å/ns, and following the instantaneous displacement, two key indicators of system relaxation were tracked over a 10 ns observation window:

- 1. The number of water molecules identified as being in the ice phase.
- 2. The potential energy (PE) of all water molecules.



Figure S4: (a) Simulation setup used for quantifying the relaxation time of water molecules following an instantaneous perturbation. Unlike the main simulations, the lower substrate undergoes a rapid displacement (pulling speed of 500 Å/ns) equivalent to the vibration amplitude and then remains stationary. Subsequent behaviour of the confined water molecules is analysed. (b) Temporal evolution of the number of ice-phase water molecules following the sudden displacement. (c) Corresponding temporal evolution of the potential energy (PE) for all water molecules, with exponential fits yielding an average relaxation time $\bar{\tau}_r = 3.37$ ns.

The temporal variation of these two quantities for different amplitudes (ranging from 10 Å to 20 Å) is shown in Figures S4(b) and S4(c), respectively. The potential energy data were analyzed by fitting an exponential decay model defined as:

$$y(t) = A \cdot e^{-t/\tau} + C, \tag{6}$$

where τ represents the characteristic relaxation time constant. Using the standard 5% settling criterion from control theory², we defined the total relaxation (or settling) time as 3τ , representing the duration required for the system to achieve within 95% of its equilibrium state. From these exponential fits, the extracted relaxation times consistently fell within a narrow range of 3–3.5 ns, yielding an average relaxation time $\bar{\tau}_r = 3.37$ ns (standard error = 0.19 ns). Given that effective nucleation must occur within half of an oscillation cycle (beyond this, the surface reverses direction, compressing the water and inhibiting nucleation), the relevant timescale for the nucleation saturation is twice this relaxation duration, or approximately 6.74 ns. This closely aligns with the previously observed 6.67 ns saturation timescale, validating our hypothesis.

This result provides a clear physical interpretation of the observed frequency threshold: if the surface vibrates at frequencies higher than the inverse of this relaxation time (i.e., greater than approximately 150 MHz), the AW perturbation frequency is too rapid to allow the water molecules to adequately relax back to their metastable state, thus suppressing nucleation enhancement and resulting in the observed saturation behavior.



S5 Cluster analysis

Figure S5: (a) Detected cluster of ice phase molecules superimposed on the molecular trajectories. (b) Flow chart of the cluster analysis conducted at each timestep

As mentioned in the paper (Section 3.3), we track the temporal evolution and dissolution of clusters of molecules in the ice phase (shown in Figure S5(a)) to analyze the influence of surface vibration on ice nucleation. This is achieved by identifying and tracking neighbors using a distance-based criteria described below (Figure S5(b)):

A) Determination of ice molecules: We employ the CHILL+ algorithm³ through OVITO⁴, which uses the number of staggered and eclipsed water–water bonds to identify water molecules in one of the ice phases (cubic ice, hexagonal ice, or interfacial ice).

B) Identification of clusters: A cluster is defined as a group of ice-like molecules where each molecule is directly or indirectly connected to at least one other molecule in the group, with connections established by being within a specified cutoff distance of 3.2 Å^5 . A direct connection occurs when two molecules are neighbors within the cutoff distance, while an indirect connection links molecules via a chain of neighbors, forming a continuous cluster (see Figure S5(a)). Molecules not connected to any others within the cutoff distance are considered isolated and are not part of any cluster.

C) Identification of clusters above the threshold size: It was observed that the cluster sizes vary widely. Smaller clusters tended to dissolve easily during the upward motion of the surface, while larger clusters were more likely to persist. To account for significant clusters only, we arbitrarily set a threshold size of 25 molecules and tracked clusters of this size or larger.

D) Tracking clusters close to the surface: Since nucleation primarily initiates on the surface, only clusters within a distance of 10 Å from the surface are considered. Clusters farther from the surface are screened out to minimize noise in Fig. 4 in the main paper.

E) Recording cluster information: Once the clusters of interest are identified, the coordinates of their center of mass are determined. Both the center of mass and the size of the clusters are recorded from configurations saved at predefined intervals leading up to nucleation.

S6 Bulk water pressure measurements



Figure S6: Variation of bulk water pressure along the direction of surface vibration as a function of time.

To verify whether the pressure in the water region away from the surfaces is consistent with P_{loc} as discussed in Section 3.4 of the paper, we also estimated the bulk water pressure. This is determined from the diagonal components of the per-atom stress tensor, specifically the normal stress (accounting for both kinetic and potential contributions) along the xx-direction. These components are summed across all atoms in the system to capture the overall stress. The bulk pressure is then calculated by averaging the summed values and normalizing by the volume of the simulation box. Figure S6 shows the variation of P_w as a function of time. It can be observed that P_w exhibits the same oscillatory behavior as P_{loc} , with approximately the same phase but slightly lower magnitude.

S7 Influence of temperature fluctuation on nucleation

In the present study, nucleation is observed under nonequilibrium conditions in which both temperature and pressure undergo continuous temporal fluctuation, which is challenging. As Figure S1(a) showed, there is a small variation in temperature during the surface vibration process. This could potentially be significant given the well-documented exponential dependence of nucleation rates on temperature. As the volume of water undergoes periodic contraction and expansion due to surface vibration, one might intuitively hypothesise that liquid expansion converts internal energy from kinetic to potential energy, resulting in a temperature reduction that facilitates ice nucleation, which would



Figure S7: Energy interactions for a representative case with amplitude = 12 Å and frequency = 100 MHz. (a) Variation of the temperature of water molecules (blue line) and displacement of the surface (red line) over time during a complete oscillation cycle. Data from the first 10 oscillations are analysed. The first cycle exhibits slightly different characteristics due to initial transience; from the second cycle onward, the fluctuations become consistent. Therefore, data are averaged over 9 oscillation cycles and presented in dark blue, while the first oscillation is shown separately in translucent light blue. (b) Variation of kinetic and potential energy as a function of time (averaged over 9 cycles; the first cycle is shown separately using a translucent line). (c) Temporal variation of the kinetic energy of water molecules, the force exerted by water molecules on the surface, the instantaneous work done by the surface, and the cumulative work. The time points T/4, T/2, 3T/4, and T (T = time period of oscillation) are indicated by vertical red lines. (d) Variation of ice-phase water molecules (blue line, interfacial ice is included here while not in the main paper) and temperature (red line) over 10 oscillation cycles. (e) Potential energy change of ice-phase molecules at each temperature peak (relative to the start of the oscillation) vs the corresponding change in kinetic energy of all water molecules.

contradict the findings of this paper.

However, our additional investigations rule out any such temperature-driven nucleation effects. The temperature remains slightly *elevated* (rather than *lowered*) relative to the initial thermostat setting for the majority of each oscillation cycle. As a result, nucleation is observed to occur at temperatures higher than the initial value, and not lower. Figure S7(a) presents the temporal variation of temperature during a complete oscillation cycle for a representative case (amplitude = 12 Å, frequency = 100 MHz), along with the corresponding surface displacement. As described earlier, nucleation is associated with the downward motion of the surface; the relevant temporal window is shaded in green. Notably, nucleation is observed to occur near the temperature peak. This observation strongly suggests that temperature fluctuation does not facilitate nucleation.

Furthermore, as shown in Figure S7(b), the variation in kinetic energy due to temperature change is considerably smaller than the variation in potential energy. This implies that pressure variation — as reflected in changes in potential energy — plays a more dominant role in the process. While the cooling of an expanding molecular system is characteristic of adiabatic conditions with constant internal energy, such a scenario is not applicable here. The system under consideration is not adiabatic, due to the presence of constant-temperature walls, and the vibrating surface performs work on the fluid during each oscillation, thereby inducing periodic changes in internal energy. Figure S7(c) displays the temporal variation in the force exerted by water molecules on the surface, the instantaneous work done by the surface, the cumulative work, along with the kinetic energy of the system. The time points T/4, T/2, 3T/4, and T, where T is the oscillation period, are indicated by vertical red lines. Here, positive force denotes repulsion and negative force denotes attraction. If surface displacement occurs in opposition to the direction of the force, the instantaneous work is considered positive (work done on the system); if surface displacement is in the same direction as the force, the work is negative (work done by the system). At the onset of each oscillation, the surface moves upward against a repulsive force, performing positive work and thereby increasing the internal energy of the system, which in turn raises both kinetic and potential energies. This continues until the surface reaches the topmost point. As it reverses direction, moving downward along the force vector, the system performs work and loses internal energy, markedly altering the slope of the kinetic energy curve and producing a characteristic peak. Although the kinetic energy decreases, the reduction is relatively minor, as the system predominantly utilises its potential energy during this phase. Beyond T/2, the force becomes attractive. The continued downward motion, now opposing the force direction, again results in positive work being done on the system. This increases both cumulative work and kinetic energy. After 3T/4, the upward movement of the surface aligns with the attractive force, causing the system to perform work and thus lose internal energy, resulting in a reduction in temperature.

Although the interchange of kinetic and potential energy in response to surface vibration is complex, the variation in kinetic energy may be attributed primarily to two mechanisms: (i) the release of latent heat (i.e. enthalpy of phase change) during a first-order phase transition, and (ii) dissipation of energy due to irreversibility in the system through mechanisms such as viscosity. The former is particularly relevant here. As shown in Figure 3 in the main article, the number of water molecules in the ice phase fluctuates throughout the oscillation, leading to periodic enthalpy release and absorption associated with phase change.

This is further examined in Figure S7(d), which compares the oscillation of temperature with the fluctuation in the number of ice-phase water molecules. The two quantities exhibit strong phase alignment. To quantify the contribution of phase transition to temperature variation, the potential energy change of ice-phase molecules at each temperature peak is computed relative to the start of the oscillation, and compared with the corresponding change in kinetic energy of all water molecules (Figure S7(e)). The horizontal axis denotes potential energy change of the ice-phase molecules; the vertical axis, the kinetic energy change of all the water molecules. The diagonal line represents situations where the changes are equal (both axes are equally scaled). The fact that all markers fall below the diagonal indicate that enthalpy release due to ice formation exceeds the rise in kinetic energy. These findings confirm that temperature fluctuations are driven primarily by phase-transition energetics, rather than acting as a precursor to nucleation. It is also important to consider the role of the constant-temperature walls, which exchange heat with the system throughout the oscillation cycle, absorbing or releasing excess energy (i.e. the difference between the potential and kinetic energy changes). Dissipative mechanisms due to irreversibility, although present, operate on longer timescales. This is evident in Figure S7(c), where the average slope of the cumulative energy curve indicates the rate of dissipation due to irreversibility, while the much steeper instantaneous slopes correspond to rapid energy fluctuations in each cycle. Thus, enthalpy release due to phase change emerges as the principal driver of temperature variation in this system.

S8 Impact of cavitation on ice nucleation

The key finding of the manuscript is that the nucleation is observed as a result of the monotonic increase in the magnitude of negative pressure with increasing strain induced by the moving bottom surface. This negative pressure continues to intensify until reaching the threshold necessary for ice nucleation. However, the occurrence of cavitation (defined as the formation of vapor cavities or bubbles when the tensile stress in the fluid exceeds intermolecular cohesive forces) introduces a different physical mechanism. As soon as cavitation initiates, the newly formed vapor cavity acts as an immediate pressure-release mechanism. Vapour cavities are highly compressible structures, rapidly equilibrating the negative pressure to near-zero values. Despite subsequent increases in surface displacement amplitude (and thus volumetric strain), the existence of the vapor cavity prevents further buildup of negative pressure, as the additional strain energy is absorbed by the expansion of the cavity rather than further lowering the pressure within the surrounding liquid.

This rapid pressure equilibration explains the observed suppression of further ice nucleation enhancement after the onset of cavitation. This mechanism is illustrated in Figure S8 for a representative case (f = 200 MHz, A = 26 Å). Upon bubble formation at roughly 3.5 ns into the simulation, the negative pressure abruptly relaxes to a value close to 0 MPa, demonstrating the immediate pressure equalization effect caused by the cavitation event (the inset shows the bubble location). Note that the regime in which nucleation occurs with cavitation in the regime map, this is likely because the nucleation occurs before the pressure can be equalised by the cavitating bubble, and therefore the nucleation event occurs alongside cavitation.



Figure S8: Variation of pressure with time, where the pressure initially tracks the surface position as expected. The formation of a vapor bubble at 3.5 ns due to cavitation, however, leads to rapid equilibration of negative pressure within the liquid. This immediate pressure release prevents further pressure-driven enhancement of ice nucleation despite continuing increases in amplitude and strain.

S9 Effect of confinement

In the current configuration, the water layer trapped between two surfaces could give rise to confinement effects, which may cause significant changes in the physical, structural, and dynamical properties of the water due to spatial restrictions imposed by the close proximity of the surfaces. These effects arise because the liquid molecules experience interactions with the confining surfaces in addition to their intrinsic intermolecular forces. Therefore, it is essential to ensure that such effects are not present in the configuration so that it represents a scenario similar to nanopores at larger scales, allowing the results to be upscaled well beyond the scale considered in the current study. We analyzed the local density of the water layer to ensure that no confinement effect is present at any scale in this study. The density was determined based on the coordinates of all water molecules, assuming axisymmetry around a centroidal axis perpendicular to the solid surface. The entire water domain was divided into cells, each measuring 0.1 Å in size, and the local mass density for the setup with the thinnest water layer employed in the current study. A closer inspection of the figure reveals two peaks at each end, corresponding to the quasi-liquid



Figure S9: Density distribution along the axis perpendicular to the solid surface for a water layer with a thickness of 100 Å.

layers adjacent to the surfaces⁶. Between the quasi-liquid layers on both surfaces, the density of the water converges to approximately 1 g/cm^3 at the center, which is consistent with the density of bulk water. The bulk-water-like density implies that the surfaces do not interfere with the water away from the surfaces. Given that this phenomenon is observed for the case with the thinnest water layer, we can confirm that all other systems with larger water thicknesses will exhibit a similar density profile as shown in Figure S9, thereby excluding the confinement effect.

S10 Bulk modulus of water

As mentioned in the main paper, a separate simulation is carried out to determine the stress-strain relationship of supercooled water at the same temperature (220 K) as in the vibration simulations. The simulation domain consists of a periodic box containing 19,317 water molecules. Initially, the temperature of the water is kept at 290 K under an external pressure of 0 bar. During the first timestep of the simulation, the temperature is reduced to 220 K over 3 ns while keeping the pressure constant, using a Nosé–Hoover thermostat and barostat with a timestep of 1 fs. The system is subsequently equilibrated for an additional 1 ns in an *NPT* ensemble (temperature = 220 K, external pressure = 0 bar). After equilibration, the water box reaches a size of $83.8 \times 83.8 \times 82.6 \text{ Å}^3$. Following equilibration, deformation simulations are conducted. A constant strain rate of 10^7 s^{-1} is applied along the *x*-, *y*-, and *z*-dimensions over a period of 10 ns to expand the water box. The stress components σ_{xx} , σ_{yy} , σ_{zz} in each dimension are calculated from the



Figure S10: Variation of σ with ε for bulk supercooled water at 220 K.

corresponding stress tensors. The volumetric stress σ is then computed as

$$\sigma = \frac{1}{3}(\sigma_{xx} + \sigma_{yy} + \sigma_{zz}),\tag{7}$$

while the volumetric strain (ε) is calculated as

$$\varepsilon = (1 + \varepsilon_{xx})(1 + \varepsilon_{yy})(1 + \varepsilon_{zz}) - 1, \qquad (8)$$

where ε_{xx} , ε_{yy} , and ε_{zz} are the strain components along the x-, y-, and z-directions, respectively. Figure S10 shows the variation of σ as a function of ε , as obtained from the simulation. It can be reasonably assumed from the figure that the volumetric stress σ is linearly dependent on the volumetric strain ε within the pressure range encountered by supercooled water in the vibration simulations, such that

$$K_w = -\frac{\sigma}{\varepsilon}.$$
(9)

where K_w is the bulk modulus of water. Therefore, by performing a linear fit to the data, the bulk modulus K_w is calculated to be 3.16 GPa, consistent with the experimentally measured value⁷.

S11 Effect of water model on regime map

All simulations in the paper were conducted using the standard mW model. However, it has been shown that this model does not accurately capture the well-known density anomaly of water, potentially impacting its quantitative accuracy for nucleation studies



Figure S11: (a) Temperature ramp simulation of ML-mW water from 290K to 200K (top panel) and the corresponding potential energy changes (bottom panel). The nucleation temperature is identified as the temperature at which potential energy sharply decreases. (b) Representative case studies conducted with ML-mW water, overlaid on the regime map from the original paper. Circles indicate results consistent with the original mW simulations, confirming the robustness of the nucleation regimes. Regime names reflect the number of realizations nucleating: complete nucleation (3/3), moderate nucleation (2/3), rare nucleation (1/3), and no nucleation (0/3).

under negative pressure. Specifically, modified versions of the mW model (e.g., the MLmW model developed by Chan et al.⁸ and utilized by Rosky et al.⁹) have demonstrated improved representation of this anomaly, particularly relevant under conditions of negative pressure. To explicitly address this limitation, we performed additional validation simulations using the ML-mW model. We selected representative cases corresponding to each of the nucleation regimes identified in our original regime map (complete nucleation, moderate nucleation, rare nucleation, and no nucleation). Each regime was represented by a single parameter combination (amplitude and frequency) from the original study, and three independent realizations were conducted for each parameter set to ensure statistical reliability.

Figure S11(a) demonstrates the temperature ramp simulations conducted from 290 K down to 200 K, using ML-mW water. The corresponding potential energy profile reveals a characteristic sharp drop upon nucleation, clearly identifying the nucleation temperature. The results from these simulations, summarized in Figure S11(b), confirm that the ML-mW model qualitatively reproduces the same nucleation regimes discovered in the original mW simulations:

- Complete ice nucleation (green circles): all three realizations exhibited nucleation.
- Moderate ice nucleation (light green circles): two of three realizations nucleated.
- Rare ice nucleation (white circles): only one of three realizations nucleated.

• No ice nucleation (red circles): none of the three realizations nucleated.

These ML-mW results validate the overall structure and boundaries of the nucleation regimes reported in our main manuscript. While some minor quantitative shifts in the exact locations of regime boundaries might emerge if the full parameter set were re-examined with the ML-mW model (which is computationally prohibitive), the demonstrated consistency across selected parameter sets strongly supports the robustness and generalizability of our findings. It confirms that our conclusions regarding the fundamental physical mechanisms of AW-induced ice nucleation are insensitive to the specific choice of water model, provided the essential physics (tetrahedral coordination and negative pressure response) is reasonably captured.

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