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Experimental Study of Frost Crystals Dendrite Growth on Two Neighboring Separate Frozen Water Drops on a Cryogenic Cold Surface under Natural Convection Conditions

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Abstract: The effects of cold surface temperature, wet air state (temperature and humidity) and original drop size on frost dendrites growth of two neighboring separate frozen water drops of same size under natural convection conditions were investigated by quantitative measurement. It was determined that for different cold plate surface temperature conditions, i.e., the ordinary-low temperature and the cryogenic temperature range, the frost formation mechanism is different. Under the conditions that the air temperature is not too high and absolute humidity is not too excessive, the influence of frozen water drop size on the longest dendrite of frost crystals becomes more and more obvious with the decrease in cold plate temperature. The changes in air temperature and relative humidity both change air absolute humidity, so they have similar effects on the growth of dendrites. However, the effect of wet air state on the growth of frost dendrites is not monotonous, which needs to be considered comprehensively in combination with heat and mass transfer and the existence of heavy phase layer. The thickness of 'the initial continuous frost layer' was measured and it was disclosed that the initial frost layer thickness is 1.7–3.0 times that of the height of the frozen water drop diameter. This value may be possibly used as initial frost layer thickness in heat and mass transfer-based frost layer growth prediction models, at least for ordinary-low temperature conditions.

Keywords: dendrite growth; frost crystal; initial frost layer thickness; cryogenic cold surface

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1. Introduction

The frost deposition is a process involving heat and mass transfer, phase transition dynamics, crystal growth physics, and moving boundaries in porous media. Hayashi et al. [1] is the first to divide the frost layer growth into three typical stages, i.e., the frost crystal growth period, the frost layer growth period and the frost layer full growth period. As successful as the frost formation research can be, the first stage is everything but fully understood. The main difficulty lies in the direct observation due to its very short duration and complex physics of the crystal growth period. Most of the experimental and simulation studies on frost formation are in the second and the third stage under ordinary low temperature conditions, i.e., the frost layer growth period and the frost layer full growth period, not to mention the frost formation on ultra-low temperature cold surfaces [2–4].

In solving various heat and mass transfer-based frost growth models, the initial frost thickness has to be provided. This is because this kind of model is based on the assumption that the porous frost layer is continuous. However, in the very early stage, at least in the nucleation stage, this assumption is not valid because only separate and independent frost nuclei exist in this stage. Therefore, it is not surprising that researchers have long noticed that the solution of these models is very sensitive to this initial value and a slight change in the initial frost thickness value may lead to a dramatic change in the prediction results. Liu et al. [5] seems to be the first who established a complete heat and mass transfer mathematical model of frost formation on a vertical cold plate under natural convection conditions that considered frost density and temperature nonlinear variations along the frost thickness direction. Jones and Parker [6] determined that if the initial thickness is chosen to be close to zero (~2×10^{-5} m), the initial frost density might vary over a wide range of values, somewhere between 8 and 48 kg/m^3. Their model could be applied from the time zero by using sufficiently small initial values for frost layer thickness and density. Based on that, some researchers selected initial frost thickness and density values within this range for numerical simulation [7–10]. Since the initial conditions need to consider the influence of working conditions and surface properties, Chen et al. [11] and Lee et al. [12] selected proper values from the experimental data in order to avoid the randomness of the numerical results, and more precise results were obtained. However, as one could understand, this is not practical, because experimental validation is required for every different model case. For abovementioned models, the initial frost thickness and frost layer density can be kept within a certain range; however, this is not suitable for all models; for example, the Lee's [13] model has a strong dependency on initial density conditions.

So far, a huge number of theoretical and experimental studies on the frosting phenomenon have focused on the cold surface temperature in the range of −30 °C~0 °C (ordinary-low temperatures) [14,15], and only a few of research works on frost formation on the cold surface in cryogenic temperature range have been reported [16,17]. Under ordinary-low cold surface temperature conditions, frost layer formation on cold surface first undergoes the process of water vapor condensing into water droplets, then these water droplets freeze to form initial frost crystals and finally grow dendrites to form frost layer. This is a process of vapor condensation frost formation [18–21]. However, for frosting under cryogenic cold surface temperature conditions, there are many special phenomena that are different from ordinary-low cold surface temperature conditions. Liu et al. [22] was the first to determined that when the temperature of cold surface temperature is lower than −165 °C, a layer of liquid air appears on the cold surface. The thickness of frost layer decreases with the decrease in cold surface temperature, which is different from the growth law of frost layer on ordinary-low temperature conditions. Li et al. [23] determined that the frost layer growth is controlled by outer boundary layer under ultra-low temperature conditions and there is a transition temperature that can cause a change in frost formation mechanism. Byun et al. [13] showed that under cryogenic temperature, a considerably low-density frost layer is usually formed due to the fact that the frost formation mechanism is sublimation dominant and is different from the conventional condensation freezing.

As it has been noted, the initial stage plays an important role in studying entire frosting process and cannot be ignored. However, the estimation of the thickness and density of this initial frost layer is anything but successful, because it involves complex physical phenomena though it lasts for a short period of time and is different for different cold surface temperatures. Song et al. [24] further divided the frost crystal growth period into droplet condensation stage and solidification liquid tip growth stage. To the best of the present authors' knowledge, most of the experimental studies on frost crystal growth reported in the open literature are focused on the freezing process of droplets under ordinary-low cold conditions with limited work on dendrite growth after solidification. These experiments are necessary but not sufficient, especially in cryogenic cold surface temperature range. The frost formation skips the freezing stage of water drop solidification and directly forms frost crystals attached to the cold plate which is a direct result of frost formation mechanism change under cryogenic conditions. In this case, the initial stage phenomenon could not be correctly explained by the freezing process of water drops. The main purpose of this paper is to solve the initial thickness problem of frost layer thickness prediction models, focusing on the dendrites growth after water drops are frozen. The quantitative study of dendrites growth and frost crystal height after dendrites intersection of two frozen water drops were carried out, aiming at finding out the initial thickness of a continuous frost layer that is first formed during frost formation.

2. Experimental Apparatus and Methods

Experimental System and Method

The experimental system consists of a refrigeration system, an air temperature and air humidity control system and a data acquisition system as shown in Figure 1. Liquid nitrogen is used as refrigerant and the cold test surface is set horizontally, 150 mm long, 50 mm wide and 10 mm thick. Nitrogen gas is used to drive the liquid nitrogen flow through the lower part of the copper plate in order to cool the cold plate by evaporating and maintain the expected temperature by changing the liquid nitrogen's mass flow rate. The cold surface temperature can be adjusted from −195 °C to 0 °C with the flow of liquid nitrogen that is controlled manually. The air temperature and humidity are regulated by air temperature control system and ultrasonic humidifier. The data acquisition system consists of T-type thermocouples, temperature and humidity sensors and a data acquisition unit. In order to observe the frost crystal growth clearly, the photos captured by photo acquisition system are stored in computer and processed by the software Image J. More detailed information on the experimental setup can be found in our previous paper [25].

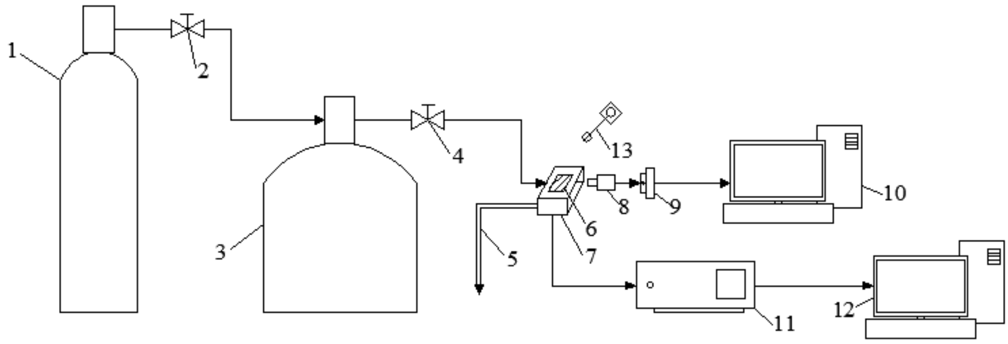


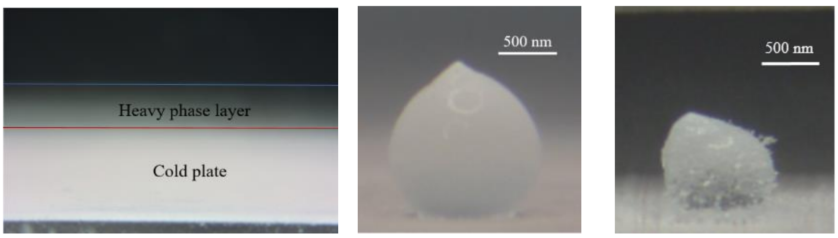
Figure 1. Experimental system. 1. Nitrogen gas bottle; 2. Intake valve; 3. Liquid nitrogen tank; 4. Inlet valve; 5. Outlet tube; 6. Experimental cold plate; 7. Tin box; 8. Microscope; 9. Camera; 10. Computer; 11. Agilent data acquisition system; 12. Computer-2; 13. Cold light source.

A plastic film was used to cover the cold plate surface before each experiment, with the central part cut off to reduce the influence of the plate edge. Another removable plastic film was applied to the exposed part to prevent wet air directly contacting the cold plate during the cooling process. The removable plastic film needs to be removed and two micro-injectors were used to quickly drip a water drop onto the naked central part of the cold plate surface when the surface temperature reached pre-set value. The longest dendrite formed on the frozen water drop and the height of the frost crystal when two frozen water drop dendrites first intersected were measured by analyzing the photographs captured from microscope camera system. The experimental temperature was divided into cryogenic temperature groups including −165 °C, −130 °C and −100 °C, and the ordinary-low temperature groups including −70 °C, −50 °C and −30 °C. The sizes of the water drops were chosen to be 0.6 µL, 0.8 µL and 1.0 µL, respectively. The height of the frost crystal after dendrites intersection and the longest dendrite were recorded and the accuracy for frost dendrite length was 0.001 mm. Image information was collected every second.

3. Results and Discussions

3.1. Frost Formation Mechanisms

It was first proposed by Holden that under ultra-low temperature conditions, frost growth occurs mainly by outer boundary layer mechanism [26]. In our experiment, as shown in Figure 2a, a fog layer is observed in the vicinity of the cold plate surface once the plastic film is removed when the cold temperature is −100 °C and lower, and we call this fog layer the heavy phase layer. The lower the temperature, the more apparent it is that this thin heavy phase layer is possibly composed of tiny liquid water droplets and/or ice particles. The existence of this heavy phase layer can be used to explain the change in frost formation mechanisms under cryogenic temperature conditions [25]. In fact, there are two conjectures about the frost formation mechanism under cryogenic temperature conditions. One is that the frost deposition could occur due to the water vapour in the air diffusing directly onto the surface and experiencing a gas–solid phase transition to form frost crystals, i.e., desublimation, Another is boundary-layer desublimation: the water vapour in concentration boundary layer is directly frozen into tiny solid ice droplets by sublimation. These tiny particles immigrate and attach to the surface to form frost crystals, like snow. Figure 2b,c presents the growth of the two frozen water drops of 2.0 µL and 0.6 µL that have contacted the cold plate for 20 s. It can be seen from the picture that frost particles deposited on the surface of the 2 µL frozen water drop, resulting in a relatively smooth surface. However, for the 0.6 µL frozen water drop, many protrusions had formed on its surface within a short period of time after freezing. The protrusions on the frozen water drop surface usually do not develop into dendrites, their appearance indicates that there are tiny liquid water droplets in the heavy phase layer and that the growth of frost crystals is strongly affected by the heavy phase layer, especially for small frozen water drops. Only freezing liquid water droplets on the frozen water drop surface can form protrusions in a short time, because otherwise, a thin frost particle layer will cover the frozen water drop surface as in the case shown in Figure 2b. It should be pointed out that the case shown in Figure 2c appears randomly for the cold plate surface temperature below −100 °C. As the water drop is freezing, the drop surface quickly forms a thin ice layer though the freezing front that grows upwards. When the temperature of the cold plate is lower than −100 °C, some dendrites will grow rapidly on the ice layer, but these dendrites are attached to the surface of the thin ice layer with only very weak adhesion. With the upward growth of the freezing front, these dendrites may fall or remain on the surface of the frozen drops quite randomly. Figure 3 is an illustration of the heavy phase layer including the tiny ice particles and liquid water droplets. The number of liquid water droplets should be less than the number of ice particles in the heavy phase layer, because if the number of liquid water droplets is greater than that of ice particles, the frozen water drop should be full of prominent dendrites under low temperature conditions, which is contrary to the experimental phenomenon.



(**a**) Heavy phase layer (**b**) 2.0 µ (**c**) 0.6 µL

Figure 2. Observed experimental phenomenon (T_w = −130 °C, T_a = 20 °C, φ = 30%).

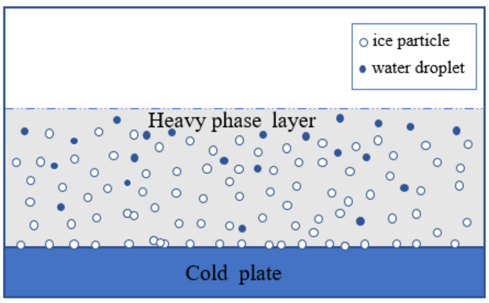


Figure 3. Illustration of heavy phase layer.

3.2. Dendrite Growth Control Equation

When the surface of a frozen water drop breaks through interfacial stability, dendrites begin to grow. In this process, the change in temperature will lead to the change in dendrite diameter. The dendrite diameter is differently formed in the different position, since the surface temperature of frost crystal is not uniformly distributed. This can explain why various-diameter dendrites appear. This section mainly deals with the theoretical relation between the dendrite diameter change and the temperature change. The detailed geometrical model is shown in Figure 4, where arrows represent heat flow density vector. It is assumed that the dendrite tip grows in a cylindrical shape of the same diameter and its solid–air interface is a plane; regardless of the dynamic effect of crystal growth, the solid–air interface is an isothermal surface with a constant temperature of water freezing point. In the process of crystal growth, the only heat source inside the closed cylindrical surface is the latent heat of solidification. If the latent heat of solidification is L and the mass of the crystal growing in unit time is m, then the heat generated in the closed curved surface in unit time Q_1 is mL. Since the solid–air interface is a plane and the temperature gradient vector is perpendicular to this plane, there is no heat flow on the cylindrical surface of the closed surface. The heat only flows along the normal direction of the upper and lower bottom of the column, so the net heat flowing out of the closed cylinder Q_2 is

$$-Q_2 = A\lambda_s \frac{\partial T_s}{\partial z} - A\lambda_g \frac{\partial T_g}{\partial z}. \quad (1)$$

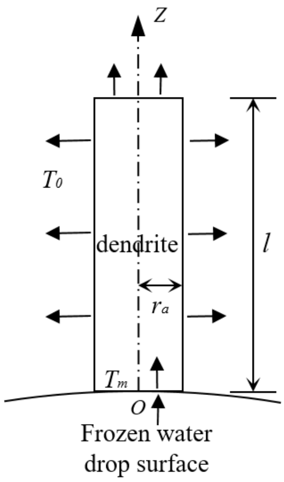


Figure 4. Geometrical model for dendrite temperature field.

Therefore, it can be concluded that the energy conservation equation at the solid–gas interface is

$$Lm = A\lambda_s \frac{\partial T_s}{\partial z} - A\lambda_g \frac{\partial T_g}{\partial z}. \quad (2)$$

When the mass of crystal accumulated in unit time $m = AV\rho_s$ is substituted, one can obtain

$$V\rho_S L = \lambda_s \frac{\partial T_s}{\partial z} - \lambda_g \frac{\partial T_g}{\partial z}, \quad (3)$$

where T_s, T_g, λ_s and λ_g are the temperature and the thermal conductivity of solid phase and gas phase, respectively; L is the latent heat of solidification, ρ_s is the branch crystal density, A is the cross-sectional area of the dendrite, and V represents the crystal growth rate. In order to solve Equation (3), it is approximately considered that the temperature field has cylindrical symmetry, and its axis of symmetry is the rotation axis of the crystal, and conduction is the only heat transfer mode in steady state. Thus, the problem is reduced to a two-dimensional steady-state heat conduction in cylindrical coordinate system, whose governing equation and boundary conditions are as follows:

$$\frac{1}{r}\frac{\partial T}{\partial r}+\frac{\partial^2 T}{\partial r^2}+\frac{\partial^2 T}{\partial z^2}=0. \tag{4}$$

At the interface, the temperature is supposed to be constant and equals to T_m, and the air temperature is T_0, so there is

$$T = T_m,\ z = 0. \tag{5}$$

The boundary condition at the top of the dendrite can be written as

$$-\lambda_s\frac{\partial T}{\partial z}=h(T-T_0),\ z=l, \tag{6}$$

and the boundary condition of the dendrite surface is assigned as

$$-\lambda_s\frac{\partial T}{\partial r}=h(T-T_0),\ r=r_a, \tag{7}$$

where h is the convective heat transfer coefficient between the dendrite surface and the air, and as a good approximation, it is taken as a constant. h can be estimated by the standard methods or empirical correlations recommended in various heat transfer textbooks and/or handbooks. The z = l and r = r_a represents the top surface and side surface of dendrite, respectively.

Using the above equations, Min [27] was able to relate Δd the change in dendrite diameter and ΔT the change in temperature as follows:

$$\Delta d=\frac{\Delta T}{(T_m-T_0)\sqrt{\frac{\lambda_s h d}{\lambda_g^2}}}\frac{d^2}{\delta_T}, \tag{8}$$

where δ_T is the boundary layer thickness and can be related to the convection heat transfer coefficient h_{tip} with air at the dendrite tip interface by the following approximate relation:

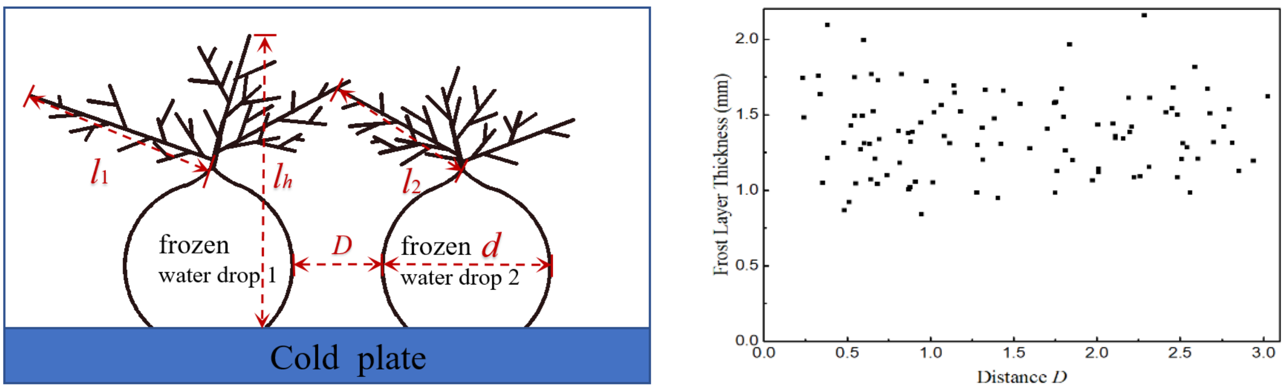
$$\delta_T=\frac{\lambda_g}{h_{tip}}. \tag{9}$$

Equation (8) presents the change in dendrite diameter caused by temperature change. From this equation, one can see that the diameter of the dendrite is very sensitive to the temperature change. A slight change in temperature will cause sharp variation in the dendrite diameter. This explains at least partly why there are so many dendrite shapes and various diameters and the great differences in dendrite shape between the cryogenic temperature and ordinary-low temperature group frost accumulation.

3.3. Frost Crystal Dendrite Growth

When a liquid water drop contacts the cold surface at low temperature, it will be frozen rapidly to form a frozen water drop whose surface is usually smooth and regular in a very short period of time. Then, some bumps appear on the surface of the frozen water drop due to the disturbance effects of certain unstable factors. It is determined that the lateral growth rate of the bump is much smaller than that of the tip direction. This is because the release of latent heat of solidification increases the air temperature around the bump and the dissipation of the latent heat is much easier at the tip of the bump flange. Therefore, the bump quickly grows into an elongated crystal which is called trunk. New bumps will also appear on the trunk in order to dissipate the released latent heat faster, and the growth of these bumps results in the formation of first, second, and third branches, namely dendrites.

At the early stage, the frost crystal dendrite grows first from the tip singularity [28]. With the time passing by, the frost crystal dendrites may grow along almost all the directions of top hemispherical space of the frozen water drop, and many dendrites appear at the same time on all the possible locations. Furthermore, due to the change in micro environment and other uncontrolled factors such as weak vibrations, melting and so on, breaking of a frost crystal dendrite may take place unpredictably. Therefore, it is almost impossible to trace the growth of all the frost crystal dendrites. To overcome this difficulty, the vertical direction growth of the frost dendrites on the two neighbouring frozen water drops were recorded. Figure 5a shows the various parameters used to characterize the frost dendrites growth. In the experiment, the two neighboring separate frozen water drops are labelled as frozen water drop 1 and frozen water drop 2, respectively. The frozen water drop diameter and the distance between the two frozen water drops are represented by d and D. l_h is the initial 'frost layer thickness'. It is defined as the vertical distance from the cold plate surface (the bottom of the frozen water drop) to the farthest extensions of frost crystal dendrites on the frozen water drop when any two of the frost dendrites from the two frozen water drops first intersect. The distance D is kept from 0.5 d to 1.0 d in the experiments because this distance is easier to achieve. In order to verify the reliability of the experimental results obtained within this distance, repeated experiments of the relationship between the distance of frozen water drops and the measured initial frost layer thickness were carried out. As it can be seen from Figure 5b, it was determined that the measured initial frost layer thickness is almost completely independent of the distance D within the region covered in our experiments. Therefore, D will not be specified hereafter, and is set between 0.5 d and 1.0 d for all the experiments throughout this paper. Here, the dendrite length l refers to the straight-line distance from the root of the dendrite to the farthest end of the frost crystal dendrite; l_1 and l_2 represent the length of the longest dendrite on frozen water drop 1 and frozen water drop 2, respectively. It should be noted that the longest frost crystal dendrite is always used in each direction of each frozen water drop, thus it may not be the same dendrite that was first recorded. Furthermore, as one may expect, the longest dendrite always grows on the outside part of the frozen water drop. In addition, there are many influence factors, including the aforementioned uncontrollable factors. Hence, in this study, we concentrate our attention only on the influences of the cold surface temperature, the volume of water drop, and wet air state (temperature and humidity).



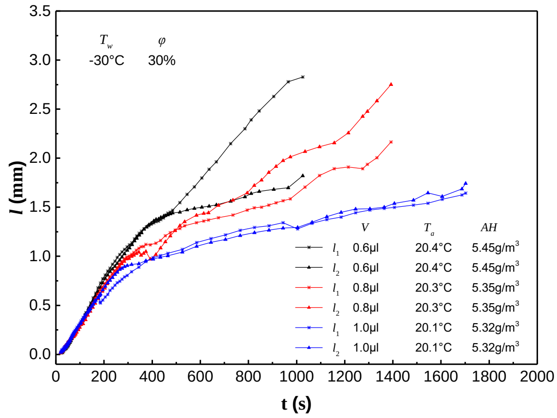
(**a**) The measurement parameters of the frost dendrites (**b**) The distance statistics between the two frozen water drops

Figure 5. Illustration of frost crystal dendrite measurement parameters.

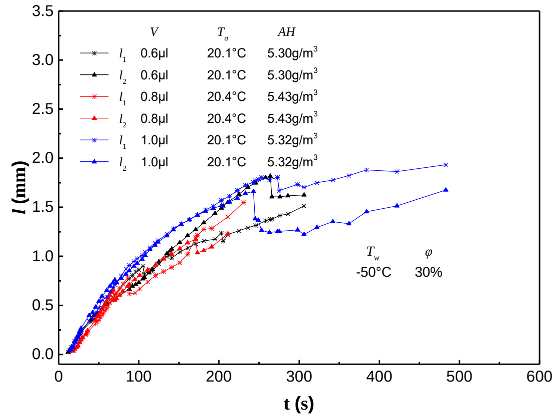
3.3.1. Influences of Cold Surface Temperature

The vertical length of frost crystal dendrite was obtained from the two-dimensional side view pictures that were captured at various instants. Figure 6 shows the function relationship between the longest dendrite and time of three different water drop sizes (0.6 µL, 0.8 µL and 1.0 µL), and six different cold plate surface temperatures (−30 °C,

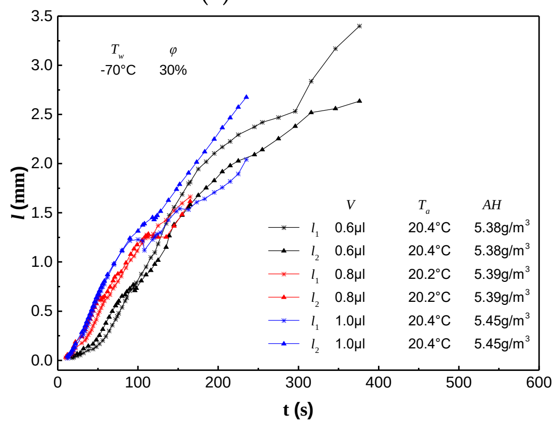
−50 °C, −70 °C, −100 °C, −130 °C and −165 °C) with an air state of temperature 20 °C and relative humidity of 30%.



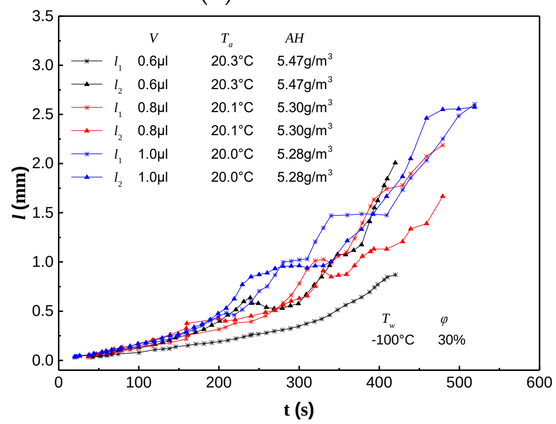
(**a**) T_w = −30 °C



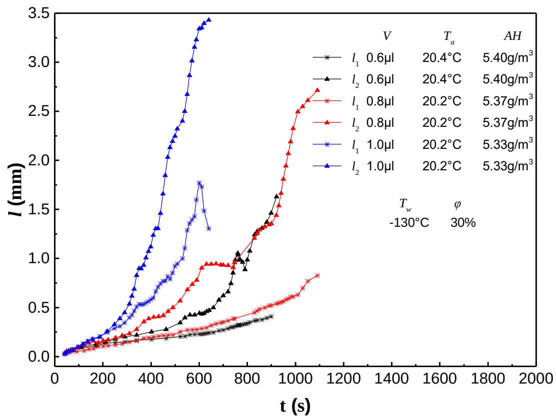
(**b**) T_w = −50 °C



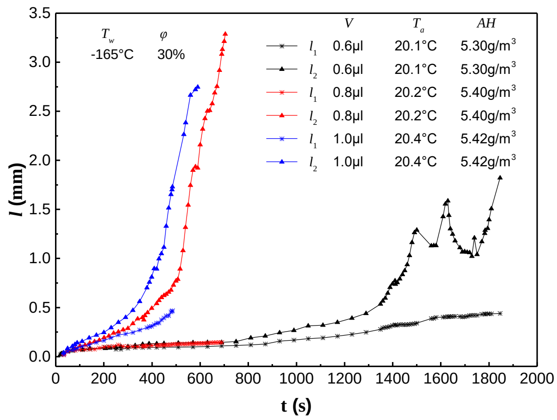
(**c**) T_w = −70 °C



(**d**) T_w = −100 °C



(**e**) T_w = −130 °C



(**f**) T_w = −165 °C

Figure 6. Influences of cold surface temperature on frost crystal dendrite growth along vertical direction.

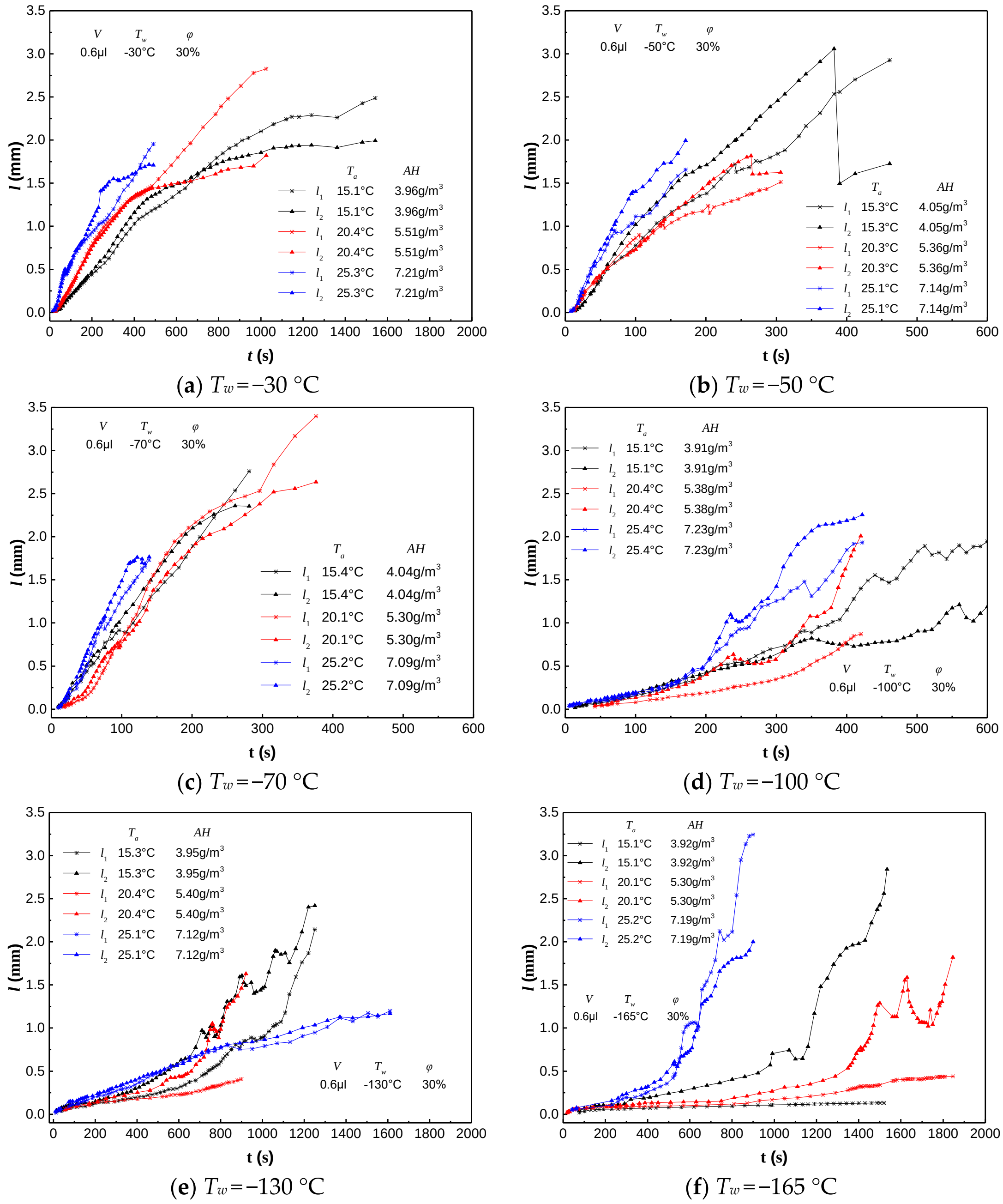
As stated earlier, the first three figures, i.e., Figure 6a–c belong to the ordinary-low temperature group; their corresponding cold plate surface temperature is −30 °C, −50 °C and −70 °C, respectively. As can be seen from Figure 6a, the frost crystal dendrites can be observed after the water droplet contacts cold plate for approximately 20 s when the cold plate surface temperature is −30 °C. The longest dendrite of three water drop sizes grows almost linearly with time for approximately 200 s, and the dendrites have similar growth rates; the corresponding length of the tracked longest frost crystal dendrite is approximately 0.7 mm. It should be noted that at the early stage, the frost crystal dendrite always grows first from the tip singularity. Dendrites broke for 1.0 μL frozen water drop at the time of 200 s, which is significantly earlier than both of the 0.8 μL and the 0.6 μL frozen water drops, whose frost dendrites broke at 400 s and 460 s, respectively. It can be seen from the case of the −30 °C cold plate surface temperature that the smaller the water drop size, the longer the time to maintain linear growth. In the first 200 s, the effect of frost crystal size on dendrite growth is not obvious, but after that, the dendrite growth rate decreases with the increase in frozen water drop size. According to heat conduction theory, as the frozen water drop size increases, its surface temperature decreases. For ordinary-low cold surface temperature, the crystal growth theory tells us that the frost dendrite growth rate decreases as cold surface temperature increases. From the above discussion, it can be concluded that lateral growth rate of the dendrites is much smaller than that of the tip direction. According to the dendrite growth control Equation (8) in Section 3.2, we know that when the temperature undercooling becomes larger, the dendrite radius increases, and the dendrite tip direction growth rate accelerates. In addition, as one can expect, the larger the frozen water drop, the earlier the frost dendrites grow into the 'far-enough' air and may thus reach ice melting point. Once the surface temperature of frost dendrites reaches ice melting point, the dendrites may melt and the dendrite growth steps into a cycle of melting growth, which also slows down the frost dendrites growth.

As it is shown in Figure 6b, the growth trend of dendrites of T_w = −50 °C is obviously different from the −30 °C cold plate surface temperature and the effect of frost crystal size on the dendrite growth rate in the early stage gradually appears. The growth rate of the frost crystal dendrites of 1.0 μL frozen water drop in the first 200 s was significantly greater than that of 0.6 μL and 0.8 μL frozen water drop. The dendrite length of the 0.8 μL frozen water drop exceeded the 0.6 μL frozen water drop at 54 s, and after that the growth trend started to fluctuate with time. As one can see from Figure 6c, in the case of the cold surface temperature of −70 °C, the dendrite growth rate is more significantly affected by frozen water drop size and increases with the frozen water drop size. This trend also applies to Figure 6d–f. The change in frost dendrite growth pattern is mainly due to the change in frost formation mechanism. As it has been pointed out, when cold plate surface temperature is −30 °C, the water vapor in the air diffuses directly to the frost crystal surface and condenses into liquid water and then freezes gradually. In this process, heat transfer plays a major role in dendrite growth. However, when the cold plate surface temperature decreases, especially below −100 °C, the water vapor in the vicinity of the cold surface may form a heavy phase layer (containing frost particles and tiny liquid water droplets), in which the particles are directly deposited on the frost crystal surface by raining or snowing. This mechanism hinders the growth of dendrite in the early stage, as shown in Figure 6d–f. There is a significant slow growth period in the early stage of dendrite growth due to frost particles deposition, and the duration of this significant slow-growth period increases as the cold plate surface temperature decreases. The presence of the heavy phase layer causing mass transfer mechanism change plays a major role in dendrite growth for cryogenic temperature group.

We now direct our attention to Figure 6d–f, which shows frost crystal dendrite variation as a function of time for the cryogenic cold plate surface temperature group. As the frozen water drop size increases, the frost crystal dendrites appear earlier and grow faster. As shown in Figure 6d, when surface temperature of the cold plate is −100 °C, the frost crystal dendrite growth rate for the frozen water drops 1 and 2 of the 1.0 μL and the 0.8 μL frozen water drop are similar. However, for the case of the 0.6 μL frozen water drop, the dendrite growth of the frozen water drop 2 is significantly faster than that of the frozen water drop 1. When the cold plate surface temperature is lower than −130°C, the growth of frost dendrites presents two patterns. One is that one of the two frozen water drops (either frozen water drop 1 or drop 2) grows dendrites first, and the other frozen water drop either remains with no frost crystal dendrites formed or its frost crystal dendrites grow slowly until it is fully covered by the frost crystal dendrites from its counterpart. Another pattern is both frozen water drops growing dendrites. The only condition for this pattern to occur is two frozen water drops developing frost crystal dendrites at almost the same time such as shown in Figure 6e by the 1.0 µL frozen water drop. It is determined in our experiments that the first pattern is the main growth state of the frost crystal dendrite growth at cryogenic temperature, as shown in Figure 6f; the growth of frost crystal dendrites on one frozen water drop inhibits the growth of its neighbouring frozen water drop. This can also explain why isolated frost crystals appear on the flat plate under ultra-low temperature conditions.

3.3.2. Influences of Air Temperature

The influences of air temperature on frost crystal dendrite growth are complicated if one considers a wide range variation of cold plate surface temperature. For the ordinary-low cold plate surface temperature, frost deposition process is usually heat-transfer controlled, i.e., as air temperature increases, the portion of the heat transfer from the cold plate surface to pre-cool air to its dew point also increases and the portion of the heat transfer for frost formation which is a phase change process from vapour to solid water is correspondingly decreased. Figure 7 displays the frost crystal dendrite growth along vertical direction as a function of time of various air and cold surface temperatures. One can observe in Figure 7a that when the cold plate temperature is −30 °C, the length of the tracked longest dendrite of frozen water drop at the time of 150 s is approximately 0.788 mm (l_1) and 0.817 mm (l_2) for 25.3 °C of the air temperature, 0.578 mm (l_1) and 0.551 mm (l_2) for 20.4 °C and 0.329 mm (l_1) and 0.34 mm (l_2) for 15.1 °C, respectively. The frost crystal dendrite growth rate is almost similar for the same size frozen water drop and has obvious differences for different-sized frozen water drops at the prophase of dendrite growth. The low air temperature condition is unfavorable for dendrite growth, because the temperature and the water vapor concentration difference between frost crystal surface and air are small in the case of low air temperature, i.e., small driving forces, causing slow dendrites growth rate. In the case of $T_w = -30$ °C, heat transfer plays a dominant role in frost crystal dendrite growth. When the temperature of the cold plate is −50 °C as shown in Figure 7b, this growth law still holds in the first 50 s though this regularity has been weakened. The only explanation for this is that the frost crystal dendrite growth is no longer dominated by heat transfer and the mass transfer may also have an impact because the air temperature determines the air's absolute humidity for the same relative humidity. In the case of $T_w = -70$ °C as shown in Figure 7c, mass transfer effects may take equal role, because frost dendrite growth trend is similar to the trend in Figure 7d–f of the −100 °C, the −130 °C, and the −165 °C cold plate surface temperature that the dendrite growth rate is the fastest at air temperature 25 °C compared to the slowest growth rate at 20 °C. For a given relative humidity, the absolute humidity increases with air temperature and for these low cold plate surface cases, the frost formation steps into the so-called mass transfer controlled mode. This is the reason of the dendrite growth rate being the fastest at the air temperature of 25 °C. However, when the air temperature is 15 °C and 20 °C, the frost dendrite growth exhibits a different trend: the frost crystal dendrite growth rate in the case of the air temperature of 15 °C is greater than that of the case of the air temperature of 20 °C, which seems to be opposite to the explanation of the mass transfer controlled mode. In fact, although the absolute humidity increases at the air temperature of 20 °C, due to the formation of the heavy phase layer at cryogenic temperature which will maintain for a longer time at lower air temperature, the growth of dendrites is promoted. It can be seen from Figure 7e,f that the dendrites of frozen water drops 1 and 2 develop at the same time and their growth rate is similar when the air temperature is 25 °C, because the growth driving force caused by the temperature difference and the heavy phase layer under high air temperature conditions are relatively large. The combination of these two factors is definitely beneficial for enhancing dendrite growth.



(**a**) $T_w = -30$ °C (**b**) $T_w = -50$ °C

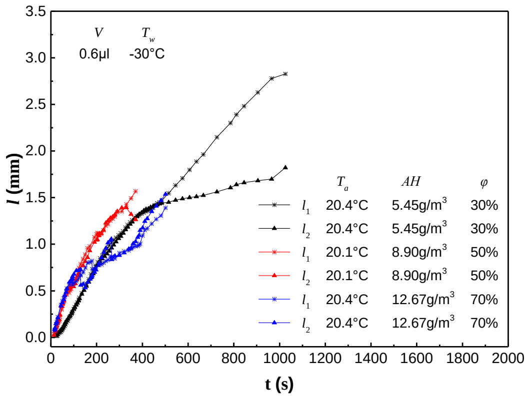
(**c**) $T_w = -70$ °C (**d**) $T_w = -100$ °C

(**e**) $T_w = -130$ °C (**f**) $T_w = -165$ °C

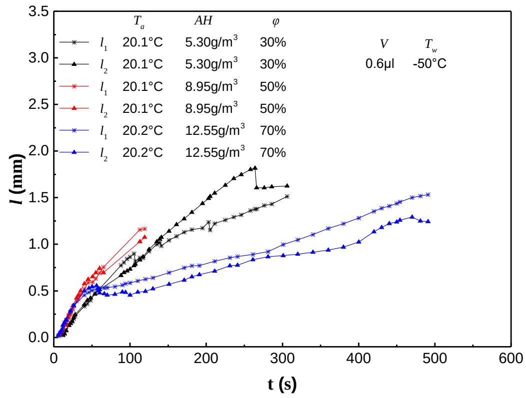
Figure 7. Influences of air temperature on frost crystal dendrite growth along vertical direction.

3.3.3. Influences of Air Humidity

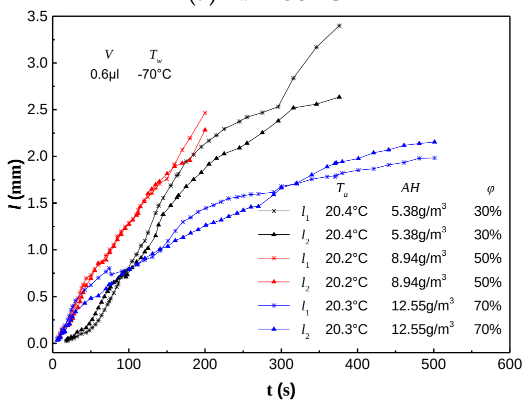
The effect of air relative humidity on the growth of the frost crystal dendrites is similar to that of air temperature at the given air relative humidity, since change in air relative humidity and air temperature changes the absolute humidity of the air. However, under the experimental conditions of this paper, the absolute humidity variation caused by the change in air humidity is significantly larger than the air temperature change, which may result in the water vapor concentration around frozen water drop surface exceed the critical value of supersaturated air concentration. Therefore, it is essential to take this fact into consideration, and it is not always true that the higher relative humidity, the more favorable to dendrite growth. As shown in Figure 8, it can be easily determined that the dendrite growth forms of ordinary low temperature group and cryogenic temperature group are significantly different. We direct our attention to Figure 8a–c. The dendrite growth trend is similar under ordinary low temperature group, since the mass transfer also has an influence under high humidity conditions compared to the abovementioned case of $T_w = -30$ °C in Figure 7a. As the cold plate surface temperature is lowered to -50 °C and -70 °C, the impact of mass transfer is gradually enhanced. Under cryogenic conditions, as shown in Figure 8d–f, the frost crystal dendrite growth is the fastest at the absolute humidity of the air is 9 g/m^3 rather than the absolute humidity of 12 g/m^3, indicating that the maximum experimental absolute humidity condition has exceeded the supersaturated concentration value. However, the mass transfer effect is enhanced again under absolute humidity conditions of 9 g/m^3 and 12 g/m^3; when the cold plate surface temperature is further decreased to -130 °C and -165 °C, the two frozen water drops both develop frost crystal dendrites at the same time and maintain similar growth rates. On the contrary, when the absolute humidity value is small, such as 5 g/m^3, only one of the neighboring frozen water drops can grow frost crystal dendrites due to the insufficient driving force for mass transfer and thus for dendrite growth.



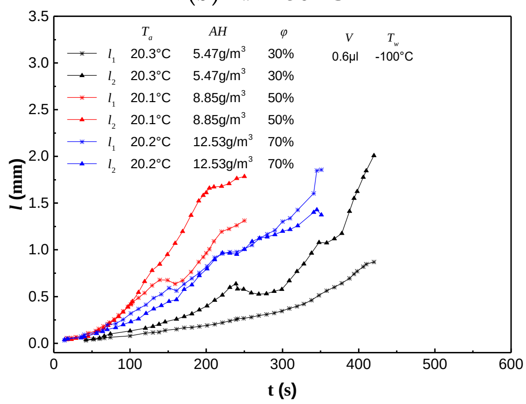
(**a**) T_w = −30 °C



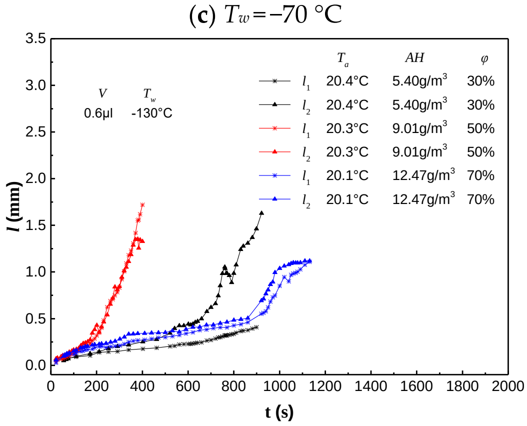
(**b**) T_w = −50 °C



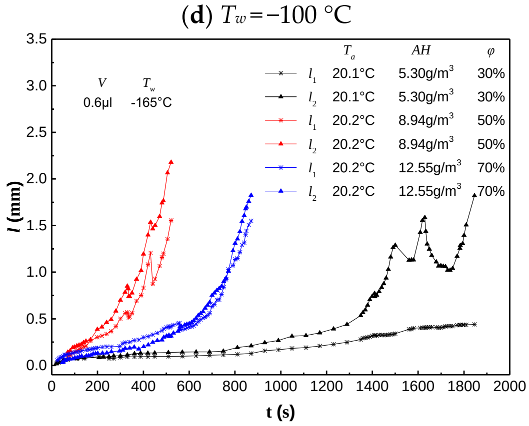
(**c**) T_w = −70 °C



(**d**) T_w = −100 °C



(**e**) T_w = −130°



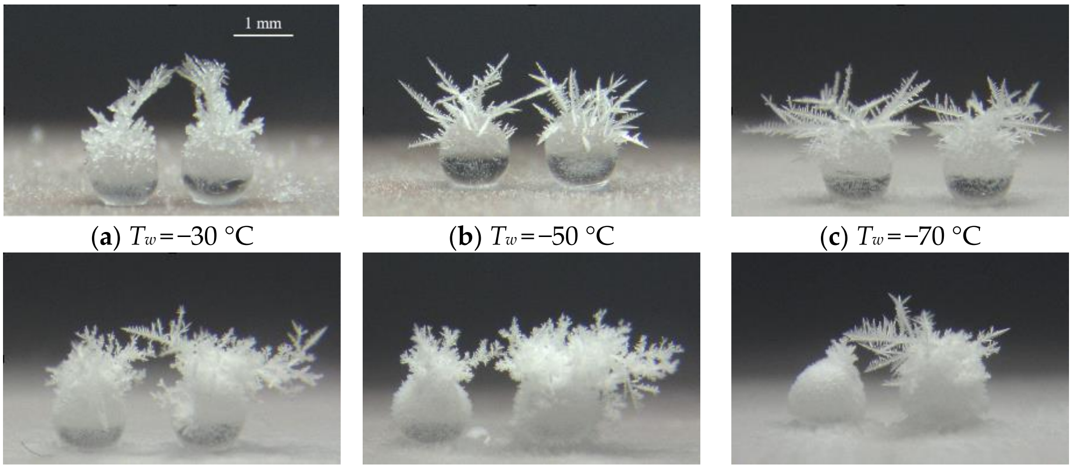
(**f**) T_w = −165 °C

Figure 8. Influences of air relative humidity temperature on frost crystal dendrite growth along vertical direction.

3.4. Initial Frost Layer Thickness

The basic purpose of this study is to analyze the dendrite growth of frost crystal dendrites at different cold plate surface temperatures, different wet air state conditions and different sizes of frozen water drops so as to provide basic information for understanding the initial stage of cold plate frosting and the growth of subsequent frost layer. What is more import is to provide a better estimation of the 'initial continuous frost layer' thickness in the initial stage of frost formation by quantitative measurements. As we all know, in the theoretical analysis and simulation study of frost formation, the thickness of the frost layer in the initial stage is needed and has been assumed quite arbitrarily. To the best of present authors' knowledge, there are no experiments reported in the literature to tackle this problem. The initial thickness of continuous frost layer is the thickness that can be treated as continuous at macro scale, which is a basic precondition for heat and mass transfer model to apply. In some research, this initial frost layer thickness was assumed to be very small, which is not correct because there is a great possibility that the 'continuous' frost layer has not been formed in any sense and it is nothing but a large number, separate and independent frost nuclei. In our experiments of cryogenic cold surface temperatures, due to the limitation of experimental means, although the size of water drops used should be close to that of nuclei formed during frost nucleation period, the realizable minimum water drop size is 0.6 μL. This is because the water drops of any smaller size will freeze before dripping to the cold plate for the cases of −130 °C and −165 °C cold plate surface temperatures. Figure 9 presents the experimental phenomena when the dendrites of two frozen water drops are just connected, i.e., the initial frost layer thickness l_h is recorded at this time. The dendrite shape and the phenomenon of forming 'continuous frost layer' are different for different cold plate surface temperatures. As can be seen from Figure 9a, when $T_w = -30$°C, the higher cold plate surface temperature means the smaller frozen water drop surface super-cooling, the dendrites melt earlier when they extend into the air. The melted water penetrates into the root of the dendrites by capillary force and induces the increase in thermal conductivity which will enhance heat conduction effect and reduce the tip temperature of the dendrites. Under this condition, melting stops and the dendrites grow again. In this condition, the dendrites formed at the tip will connect with the dendrites formed at the neighbouring frozen water drop after repeated growth–melting–regrowth process. The dendrites on the hemispherical surface of the frozen drop are difficult to extend in the air due to the small blocks formed after melting process. When the cold plate temperature decreases, as shown in Figure 9b ($T_w = -50$ ° C) and 9c ($T_w = -70$ °C), the pine dendrite state maintains for a longer time and dendrite tip do not appear to be melting even after dendrite intersection of two frozen water drops due to the increased surface undercooling. The relatively loose dendrites lead to the reduction in the density of frost layer and adhesion, which can explain the situation with the frost at these two temperatures: it does not easily adhere to the cold plate surface [23].

When the cold plate surface temperature reaches −100 °C and lower, as shown in Figure 9d–f, dendrites first appear in the form of clusters. The dendrite growth trend well confirms the frost dendrite growth patterns as we have discussed above in Section 3.3.1. The first point to consider is that one of the two frozen water drops dendrite grows will first affect the growth (slow or no growth) of dendrites of another frozen drop; the second is that both the frozen water drops grow dendrites. Under cryogenic conditions, a layer of frost particles in the very boundary region appears, and it was clearly observed, which is not detected under ordinary-low temperature conditions and confirms the desublimation frost formation mechanism [28]. The thickness of the frost particle layer (the heavy phase layer) increases as the cold plate surface temperature decreases. In fact, the observations in Figures 6–8 clearly show that the frost mechanism under ordinary-low temperature conditions is different from that under cryogenic temperature conditions. Frost is formed by condensation of water vapor under ordinary-low temperature conditions, and by desublimation under cryogenic temperature conditions of which the typical characteristic is that a layer of frost particles will be formed on the surface of frozen water drop under cryogenic temperature conditions.



(**a**) $T_w = -30$ °C (**b**) $T_w = -50$ °C (**c**) $T_w = -70$ °C

(**d**) $T_w = -100$ °C (**e**) $T_w = -130$ °C (**f**) $T_w = -165$ °C

Figure 9. Experimental phenomenon when dendrites intersect (T_a = 15 °C, φ = 30%).

Specific presentation of these two growth patterns is shown in Figure 10. Figure 10a shows frost growth process at −50 °C cold plate surface temperature with air temperature T_a = 20 °C and relative humidity φ = 30%. It can be seen from this figure that dendrites easily appear on the cold plate surface at this time, and the growth of dendrites is much larger than that of the frost nuclei, so the dendrite growth and initial thickness are little affected by the size of the frozen water drop. It is important to focus on the growth of dendrites in our experiment. When isolated large frost crystal dendrites on the cold plate surface appear, the growth of the surrounding frost crystal dendrites will be inhibited and show part of exposed area. As time elapses, the influence of the isolated large frost crystal dendrites on surrounding frost crystals dendrites will be weakened when the dendrites grow from the surrounding frost nuclei until a continuous frost layer is finally formed. Figure 10b,c is the illustration of two different dendrite growth modes when the cold surface temperature is −130 °C. As one can observe, the experimental phenomenon in Figure 10b is similar to that of Figure 10a, both of them displaying obvious isolated large frost crystals at the early stage of frost formation. However, due to the change in growth mechanism, it presents isolated clusters rather than the typical dendrite state. This isolated state gradually weakens when the large frost crystal clusters are connected with the surrounding small frost crystal clusters to form a continuous frost layer. Close observation in Figure 10c discloses that there has been no isolated frost crystal emergence, showing the second growth pattern of dendrites. The adjacent frost crystals grow in cluster dendrites at the same time with similar growth rates. A continuous frost layer state has been maintained in this case.

Table 1 lists the measured 'initial frost layer thickness' that is defined as the frost crystal height l_h when any of two frost crystal dendrites from the two separate frozen water drops (the distance D of these two drops is set at 0.5 d~1.0 d) intersect to form a 'continuous frost layer' (Figure 3). It is determined that this initial thickness is little affected by the size of the frozen water drop. As one can observe from Table 1, the maximum range of the 'frost layer thickness' after dendrite intersection is 1.756 mm~2.988 mm for the case of the 0.6 µL frozen water drop (the height of the frozen water drop of 0.6 µL without frost deposition is approximately 1.0 mm after the water drop is frozen), 1.818 mm~2.923 mm for the case of the 0.8 µL frozen water drop (the 0.8 µL frozen water drop height is approximately 1.1 mm), and 2.156 mm~3.131 mm for the case of the 1.0 µL frozen water drop (the 1.0 µL frozen water drop height is approximately 1.2 mm), respectively. Therefore, it was determined that the initial frost layer thickness defined above is roughly equal to the range of 1.7 to 3.0 times of the height of the frozen water drop under ordinary-low surface temperature conditions. This finding is important for estimating the initial frost thickness that can be simply taken as 1.7~3 times of the critical diameter of nuclei calculated according to the cold surface temperature. Although Table 1 also shows that the cold plate surface temperature has a weak influence on this initial frost layer thickness, the measurements under cryogenic temperature conditions present more uncertainty (such as no significant dendritic growth). More experiments may be needed to confirm this conclusion.

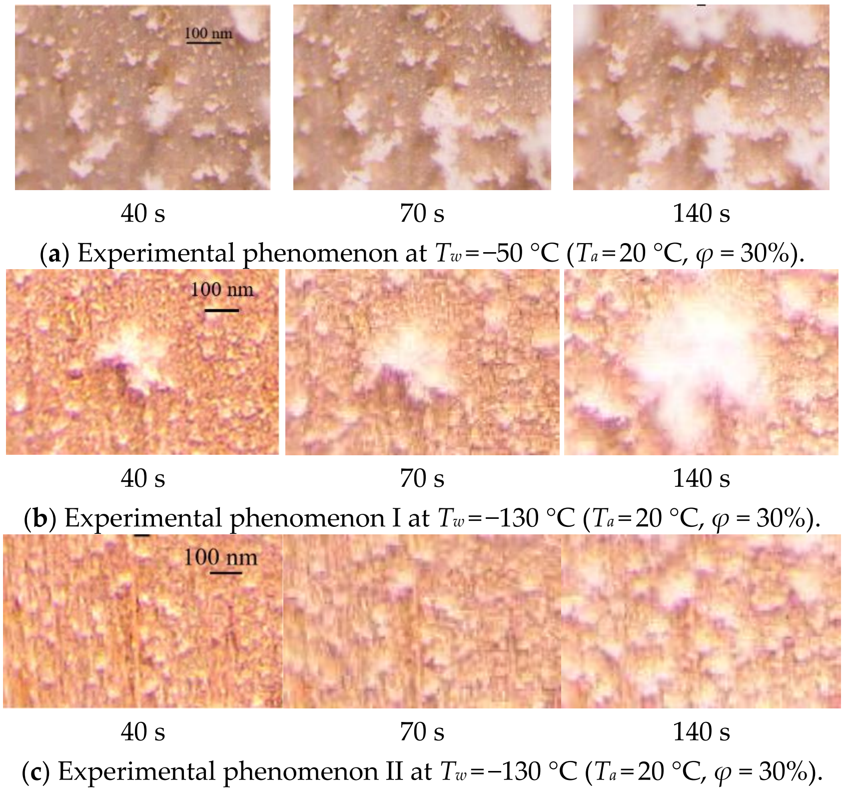


Figure 10. Experimental phenomenon of frosting at three different time instants.

Table 1. The measured initial frost layer thickness l_h (mm).

Wet Air State	V, μL	*TW*					
		−30 °C	**−50 °C**	**−70 °C**	**−100 °C**	**−130 °C**	**−165 °C**
φ = 30% T_a = 15 °C	0.6	2.848	2.243	1.972	2.988	2.706	2.393
	0.8	2.353	2.380	2.390	2.166	2.117	2.176
	1.0	2.637	2.360	2.446	2.353	2.452	3.131
φ = 30% T_a = 20 °C	0.6	2.546	2.453	2.916	2.129	1.813	1.756
	0.8	2.909	2.253	2.538	2.901	2.477	2.816
	1.0	2.739	2.993	2.878	2.469	2.237	2.549
φ = 30% T_a = 25 °C	0.6	2.242	1.902	2.330	2.390	1.837	2.162
	0.8	2.276	2.262	2.412	2.133	2.045	1.818
	1.0	2.544	2.452	2.874	2.310	2.908	2.629
φ = 50% T_a = 20 °C	0.6	2.056	2.093	2.099	2.424	2.402	2.919
	0.8	2.458	2.406	2.339	2.370	2.059	2.923
	1.0	2.296	2.156	2.760	2.339	2.991	2.581
φ = 70% T_a = 20 °C	0.6	1.892	2.137	2.413	2.164	1.922	2.123
	0.8	2.679	2.361	2.289	2.704	2.303	2.147
	1.0	2.779	2.386	2.655	2.793	2.384	2.228

4. Conclusions

Frost crystal dendrite growth on two frozen water drops has been investigated experimentally under wide range of cold surface temperature conditions, focusing on the quantitative measurement of frost crystal dendrites and the influences of cold surface temperature, initial water drop sizes and wet air states. From the results obtained and the discussions above, the following conclusions can be drawn:

(1) Quantitative measurements in the vertical direction of the frost crystal dendrites were carried out; the results showed that the length of the frost crystal dendrites on the surface of the frozen water drops increases almost linearly with time under ordinary-low temperature conditions, especially in the short period when the first frost crystal dendrite is observed. The frost crystal dendrite growth of the cryogenic temperature conditions will have a slow stage in the early period due to the appearance of frost particles. It is also determined that for most cases, the frost crystal dendrites usually prefer to grow on one of the frozen water drops, and the early and fast-growing frost dendrites from this frozen water drop may well inhibit the dendrite growth of another frozen water drop.

(2) With the decrease in cold surface temperature, the influences of the frozen water drop sizes on the longest frost crystal dendrite growth become more and more obvious. Under cryogenic temperature conditions, the dendritic growth rate increases with frozen water drop size. Under ordinary-low temperature conditions such as $T_w = -30$ °C and $T_w = -50$ °C, the dendritic growth rate is less affected by the drop size. The influences of wet air state (air temperature and relative humidity) on the growth of frost dendrites are much more complex, and may also affect the frost formation mechanism.

(3) The heavy phase layer composed of tiny frost particles and condensate droplets can be observed right above the cold plate surface when the temperature is −100 °C and lower; its existence can influence the frost mechanism. Under the experimental conditions, the longest frost crystal dendrite growth at $T_w = -100$ °C shows a different growth trend from $T_w = -70$ °C, indicating that the frost formation mechanism changes at this temperature, and the appearance of the heavy phase layer directly affects frosting. Under ordinary-low temperature conditions, heat transfer plays a leading role in dendrite growth, especially when the cold plate temperature is −30 °C, the mass transfer is enhanced with the decrease in the cold plate temperature, but the mass transfer effect is dominated due to the appearance of heavy phase layer when the cold plate temperature is −100 °C and lower.

(4) The initial 'frost layer thickness' has been determined experimentally, and it was determined that the thickness can be estimated as 1.7~3.0 times of the critical diameter of nucleation nuclei calculated according to the cold surface temperature, at least for ordinary-low temperature conditions.

The so-called initial 'frost layer thickness' obtained in this paper is strictly confined to our definition and based on the measurements of two separate frozen water drops only. This is far from true, if one considers that in reality a very large number of nuclei appears almost exactly at the same time in the initial frost formation period. Therefore, more experimental investigations are expected to clarify this problem.

Author Contributions: F.Y.: Conceptualization, Investigation, Methodology, Experiment, Writing—Original Draft; Z.L.: Conceptualization, Methodology, Writing—Review and Editing, Supervision; Y.L. (Yanxia Li): Writing—Review and Editing, Experimental, Supervision; Y.C.: Experimental; Y.L. (Yi Li): Experimental. All authors have read and agreed to the published version of the manuscript.

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Nomenclature

T_w	Cold plate temperature, °C
T_a	Air temperature, °C
φ	Air relative humidity, %
A	Dendrite cross-sectional area, m^2
AH	Air absolute humidity, g/m^3
t	Time, s
r_a	Dendrite radius, m
d	Dendrite diameter, m
m	Mass, kg
D	Distance of two frozen water drops, m
l	Length of frost crystal dendrites
l_1	Longest dendrite on frozen water drop 1, mm
l_2	Longest dendrite on frozen water drop 2, mm
l_h	Height of the frost crystal, mm
T_0	Air temperature, °C
T_m	Interfacial equilibrium temperature, °C
T_s	Temperature of solid phase, K
T_g	Temperature of gas phase, K
λ_s	Thermal conductivity of solid phase, W/(m·K)
λ_g	Thermal conductivity of gas phase, W/(m·K)
L	Latent heat of solidification, J/kg
ρ_s	Crystal density, kg/m^3
h	Convective heat transfer coefficient, $W/(m^2{\cdot}K)$
h_{tip}	Dendrite tip convection heat transfer, $W/(m^2{\cdot}K)$
δ_T	Boundary layer thickness, m

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