The freezing of raindrops falling through strong electric fields

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SUMMARY

Supercooled water drops of radius around 2 mm and temperature ranging from 0 to $-12^\circ$C were detached from a support and could be disrupted as they subsequently fell into a region of intense electric field. Photographs showed that whereas no freezing occurred if the drops were not disrupted the fraction of drops that froze increased steadily from 0 at about $-5^\circ$C to around 0.6 at $-10^\circ$C if they were disrupted. The passage of a spark to the drops during disruption did not affect the freezing probability. Studies were made on about 200 drops.

A series of experiments on bulk supercooled water and drops suspended from mechanical supports indicated that nucleation was initiated by means of cavitation. This conclusion was reinforced by high speed photographs showing the production and collapse of cavity bubbles when disruption occurred and indicating strongly that freezing initiated from the sites of these bubbles at the time of their collapse.

1. INTRODUCTION

Pruppacher (1963a) reviewed earlier work which had purported to demonstrate that supercooled-water drops can be nucleated by means of electric fields. He argued that the freezing effects observed in these studies may not be a consequence of homogeneous nucleation initiated by molecular rearrangements produced by the field but could be attributable either to freezing nuclei produced by electrical discharges, as had previously been suggested by Blanchard (1961), or to nuclei present in the laboratory air which entered the drops under the influence of the electric field. Consequently Pruppacher (1963b) performed a series of experiments in which water drops were sealed in plastic tubes containing air or silicone oil and placed in electric fields from 1 to 30 kV cm$^{-1}$. He reported that freezing was initiated at temperatures only a few degrees below 0°C in fields of several kilovolts per centimetre. He established that freezing was due neither to an orientation of molecules in the field nor to the introduction or production of freezing nuclei by the field. He noted that freezing always commenced at a triple phase interface (water-oil-plastic or water-air-plastic) and thus concluded that electrofreezing was of no consequence in the atmosphere.

Loeb (1963) analysed Pruppacher's results and deduced that, in all cases where freezing occurred, the drop was drawn out into a liquid filament by electrical forces. Thus it would seem possible for electrofreezing to occur in the atmosphere in any circumstances in which the electric field, or indeed, mechanical forces are able to produce a liquid filament. Abbas and Latham (1969a) reported that the probability of freezing of a supercooled water drop mounted on a support was greatly enhanced if the drop was disrupted by electrical or mechanical means to produce a liquid filament. The increased values of freezing probability were always obtained when disruption occurred but were never observed in the absence of disruption even when movement of the triple phase interface occurred. Similarly, the freezing probability was not increased when the droplets were subjected to an electric field just below that required to disrupt them, thereby demonstrating that the increased freezing probability was not due to material introduced or produced by the field.

The possibility that freezing may be initiated in supercooled droplets disrupted by electrical or mechanical forces may be of relevance to cloud physics in two situations. Several workers have shown recently that the concentrations of ice particles in shallow supercooled clouds located near the 0°C isotherm can be considerably in excess of the
measured concentrations of ice-forming nuclei at the cloud top. It is possible that the rapid formation of liquid filaments produced in supercooled drops by collisions or the locally intense electrical forces which will exist between closely separated drops even when the external fields are weak (Latham and Roxburgh 1966) could initiate freezing. Rough calculations showed that the minimum lifetime of a filament is greater than the time required for its freezing at temperatures below about \(-5^\circ C\). In addition, experiments have shown that the lifetimes of filaments produced by the collision and separation of drops are much longer than the freezing times. Therefore, it appears that a significant proportion of interactions producing filaments at temperatures below about \(-2\) or \(-3^\circ C\) will persist for sufficient time for a frozen drop-pair linked by an ice-bridge to be produced. Secondly, the freezing of individual raindrops disrupted by the intense electric fields in the vicinity of a lightning channel produced within a thunderstorm could give rise to the formation of a ‘hail gush’, as reported by Vonnegut and Moore (1960) and Moore et al. (1962, 1964).

The purpose of the present study was to determine whether the disruption of freely falling drops, principally by electrical forces, could initiate freezing; to establish the physical basis of this phenomenon; and to assess its meteorological relevance.

2. The freezing of individual falling drops

Preliminary experiments indicated that it was possible to shake a supercooled drop from a suitable support rod without causing it to freeze. An experiment was therefore devised in which a drop was placed upon a support, allowed to cool to the environmental temperature and shaken from the support. The drop then passed between two electrodes across which an electric field of sufficient magnitude to cause disruption of the drop was applied. Photographs were taken before and after the drop passed through the field to determine whether or not the drop was frozen.

The apparatus used in this experiment is illustrated schematically in Fig. 1. The drop was placed upon a copper rod coupled to a steel bar situated partially inside a solenoid. It was allowed to cool to the temperature of the environment, the solenoid was switched on, the support accelerated rapidly upwards and the drop was thus released. It then passed between a pair of polished rectangular electrodes 7.5 cm long, 6.5 cm wide and 2.5 cm apart, which were connected across a continuously variable 0 to 30 kV power supply.

In the first set of experiments the falling drop was photographed before and after passing through the electric field by means of two cameras and stroboscopes arranged to give ‘dark-field’ illumination. The stroboscopes were fitted with parabolic mirrors and made use of compact arc, xenon-filled discharge tubes which could be focused to give parallel beams. These were brought to a focus at the drops by means of large plano-convex lenses. The stroboscopes were fitted with stops, shown in Fig. 1, of such a size that no direct light from the optical system could enter the camera lens, the only light arriving at the cameras being that scattered by the drops. The stroboscopes were triggered by the passage of the drop through two photocell arrangements allied to timing circuits to allow the flash to be synchronized with the arrival of a drop in front of the camera.

The drops fell through a distance of 55 cm before entering the field of view of the upper camera. Calculations showed that for temperatures below about \(-2^\circ C\) the time of fall was such that a significant fraction of the drop would be frozen before it was photographed. Similarly, the time of fall of the drop from between the electrodes to the second stroboscope was large enough for the drop to be noticeably frozen by the time the second photograph was taken if the field caused nucleation.

The drops were obtained from a hypodermic syringe calibrated to \(10^{-2}\) cm³, the distilled water used having a conductivity of \(5 \times 10^{-4}\) ohm\(^{-1}\) cm\(^{-1}\). The volumes of the drops varied from 35 to 50 microlitres, corresponding to a range in equivalent radius of 2.0 to 2.3 mm. The experiments were performed in a cold room of approximately 10 m³ internal volume whose temperature could be varied from 0°C to \(-40^\circ C\).

The experimental procedure was to place a water drop of known volume upon the
support and allow it to cool for two minutes. Calculations showed that the thermal relaxation time for these supported drops was typically about 10 seconds so that the drops were in good thermal equilibrium with their environment at the time that they were detached. During this cooling period, the temperature of the air near the drop was noted. The electric field was then applied, both camera shutters were opened and the solenoid was switched on. After the drop had been photographed by both cameras the solenoid and the field were switched off and the camera shutters closed.

Measurements were made in the absence of electric fields and in the presence of fields \( F \) slightly above and slightly below the critical value for disruption, shown by Abbas and Latham (1969b) to be given for uncharged drops by the equation \( F(R/\sigma) = 1.60 \) where \( R \) is the radius and \( \sigma \) the surface tension of the drop; the applicability of this equation to these experiments was verified in a supplementary study. When the field exceeded the critical value disruptions were sometimes accompanied by a spark.

Some typical photographs produced by this technique are shown in Fig. 2. It is seen that drops were unfrozen prior to entering the field — as was found always to be the case — and frozen after disruption. It is apparent that frozen and unfrozen drops are readily distinguishable. No evidence was found in the photographs taken with the lower camera of freezing in a drop which had not been disrupted. A total of 98 photographs were taken indicating the absence of ice from undisrupted drops.
In a modified experiment designed to increase the rate of acquisition of results, the lower stroboscope was arranged to give a beam of light at 90° to the camera axis and was operated in a free-running mode; the upper stroboscope was not used. The flashing rate was such that several images of the same drop were recorded on each photograph. Typical photographs obtained with this technique are presented in Fig. 3. Since the preceding experiments had failed to provide any evidence for freezing in the absence of disruption, all the measurements with this new technique involved drops which were disrupted by the field.

A seeding process by which some of the falling drops were nucleated on passing through an ice crystal cloud demonstrated that frozen and unfrozen drops could be readily distinguished from the photographs. The unfrozen drops gave high intensity reflections of the stroboscope light in the drop surface, whereas the frozen drops provided a much more evenly illuminated image. The efficacy of this procedure is illustrated in Fig. 4. The smaller drop illustrated in Fig. 4(b) is an example of a satellite drop which was sometimes produced during detachment of the larger drop; no measurements were made of the freezing of these satellites.

The results of the experiments on the nucleation of supercooled drops by disruption in high electric fields are presented in Fig. 5. In all the cases marked as circles, disruption of the drop was accompanied by a spark discharge passing between the two electrodes and the drop. No spark occurred in the situations marked by a cross although the electric field was high enough to ensure that the drops were disrupted. It is seen that the fraction of supercooled drops that freeze after disruption in an electric field

![Figure 5](image_url)

Figure 5. Individual results showing the occurrence of freezing (above horizontal line) and no freezing (below line) for falling drops of radius 2.2 mm over a range of temperature $T$. o, spark to drop; x, no spark to drop.
Figure 2. Photographs of supercooled drops of radius 2.3 mm in free fall. A, before disruption, $-9.5^\circ C$; B, before disruption, $-13.0^\circ C$; C, after disruption, $-9.5^\circ C$, frozen; D, after disruption, $-11.0^\circ C$, frozen.
Figure 3. Falling drops of radius 2·2 mm after disruption. A, B, −7°C, unfrozen; C, −10·5°C, frozen; D, −9·3°C frozen.
Figure 4. Falling drops of radius 2.2 mm: A, unfrozen; B, frozen by artificial 'seeding' at −10°C.
increases steadily from zero at around \(-5^\circ\text{C}\) to about 60 per cent at around \(-10^\circ\text{C}\). On average about 20 drops were studied at each temperature. These values are accurate to within 10 per cent. The fraction of disrupted drops that froze was observed to be lower at each temperature than that measured for drops suspended from mechanical supports, probably because the conditions required to induce nucleation were not so favourable in the present experiments.

It may be concluded that the freezing of supercooled drops may be facilitated by disruption of the drop in an electric field and is not dependent upon the presence of a support. The passage of a spark to the drop at the time of disruption does not appear significantly to affect the nucleation process.

Experiments were also performed in which falling supercooled drops were disrupted by means of an air jet in the absence of electrical forces. Photographs obtained in the same manner as in the primary experiments provided no evidence for the presence of ice in the drops, but these studies were too cursory to be definitive.

3. The mechanism of freezing resulting from drop disruption

Four possible mechanisms by which the production of a liquid filament could result in the nucleation of the ice phase have been investigated: (i) as suggested by Mason (private communication), there may be local cooling due to the creation of new surface against the surface tension forces when the filament is drawn out from the bulk of the drop; (ii) there may be evaporative cooling of the surface of the filament due to its large surface to volume ratio; (iii) there is the possibility suggested by Loeb (1963) that molecular aggregates may be formed in the tip of the filament which serve as freezing nuclei; and (iv) freezing may result from cavitation, as suggested by Hallett (private communication). These possibilities are discussed in greater detail in the following paragraphs.

(a) Cooling due to the creation of new surface

If it is assumed that the surface energy of a liquid filament produced by the disruption of the surface of a supercooled drop is provided entirely by the abstraction of heat from the filament it is possible to calculate its maximum temperature reduction. If the filament is cylindrical, having length \(l\) and radius \(r\) then its surface energy is approximately \(2\pi rh\), where \(h\) is the surface energy/unit area. If no heat exchange occurs between the filament and either its environment or the parent drop the amount of heat required to provide the surface energy is

\[
\Delta Q = \pi r^2 \rho C_p J \Delta T \approx 2\pi rh
\]

where \(\rho\) is the density of water, \(C_p\) is its specific heat at constant pressure, \(J\) is the mechanical equivalent of heat and \(\Delta T\) is the temperature reduction produced. Rearranging Eq. (1) and inserting appropriate values of \(\rho\), \(C_p\), \(J\) and \(h\) (noting that the correct term is the 'total surface energy', defined by Tabor (1969), which is related to the isothermal surface tension \(\sigma\) by means of the equation \(h = \sigma - T d\sigma/dT\)) we obtain

\[
\Delta T(\text{C}) \approx \frac{0.06}{r}
\]

where \(r\) is measured in micrometres. We see that for filament radii of 10 \(\mu\text{m}\) and 1 \(\mu\text{m}\) we obtain values of temperature reduction \(\Delta T\) of about 6 \(\times 10^{-3}\) \(^\circ\text{C}\) and 6 \(\times 10^{-2}\) \(^\circ\text{C}\), respectively. Obviously, these values of the temperature reduction are not sufficient to provide any significant increase in the freezing probability. In order to give values of temperature reduction of 10\(^\circ\text{C}\) to 20\(^\circ\text{C}\), filaments of width not exceeding about 10\(^{-7}\) cms radius would be required. Such a filament, should it exist, would be too small for surface tension forces to govern the shape, and this theoretical treatment would no longer be valid. It appears, therefore, that this proposed mechanism is not capable of providing the required local cooling.
(b) Evaporative cooling of the filament

We consider the evaporative cooling of a cylindrical filament of radius \( r \) and length \( l \) drawn out by electrical forces from a parent drop. If we make the simplifying assumption that the filament is at a temperature \( T_f \) which is lower, because of evaporation, than that, \( T \), of the surrounding air then it can be shown that for values of \( r/l \) less than about 0.1 the exchange of heat between the filament and the parent drop is considerably less than that between the filament and the air. Since, in practice, \( r/l \) is always much less than 0.1 the influence of the drop on the cooling of the filament can be neglected and we can therefore calculate the temperature reduction \( \Delta T = (T - T_f) \) by equating the heat loss from the filament because of evaporation to the heat gain resulting from conduction through the warmer surrounding air. Following Macklin and Payne (1968) we see that this condition is achieved when

\[
\Delta T = \frac{L_v D (\rho_s - \rho_v)}{K}
\]  

(3)

where \( L_v \) is the latent heat of evaporation of water, \( D \) is the diffusion coefficient of water-vapour molecules, \( K \) is the thermal conductivity of air, \( \rho_s \) is the saturation vapour density of water at the temperature \( T_f \) and \( \rho_v \) is the density of water vapour in the air. Although the drop will ultimately undergo the temperature reduction predicted by Eq. (3) it will cool much more slowly than the filament. Substituting into Eq. (3) appropriate values of \( L_v, D \) and \( K \) we find that

\[
\Delta T(\degree C) \approx 2 \times 10^{-6} \{\rho_s(T_f) - \rho_v(T_f)\}
\]  

(4)

In the environment in which these laboratory studies were conducted the air was significantly undersaturated and Eq. (4) predicts values of \( \Delta T \) of typically about 2\degree C. However, a reduction in temperature of this amount cannot be responsible for the observed large increase in freezing probability produced by the disruption of supercooled drops. Within clouds, it is unlikely that water drops and any filaments drawn out from them will be significantly cooler than their environment because of evaporation.

(c) Reorientation of molecules in the filament tip

It is conceivable that the production of molecular aggregates in the form of minute crystallites might take place in the extremities of liquid filaments as suggested by Loeb. Although this process could, in principle, operate to induce freezing in interacting drops it is supported by such slight evidence that it should probably be regarded at the present stage as unlikely to be of major importance.

(d) Freezing induced by cavitation

When a liquid is subjected to sufficient internal stress, cavities are formed within the bulk of the liquid. The threshold stress required to form a cavity is a function of the amount of dissolved gas and also the purity of the water, since for a cavity to form, there must be a suitable nucleus in the form of a solid particle or a microbubble of gas present in the region of stress. Gavrilenko and Topchiyan (1966) have shown that very pure water can withstand internal stresses of a few atmospheres for a period of a few tens of microseconds. However, for water samples other than those of extreme purity and low gas content, cavities will normally appear if the internal pressure is locally reduced below the vapour pressure of the liquid at the temperature of the sample. Many workers have shown that cavitation may be produced by ultrasonic vibration, mechanical shocks and frictional processes. Goyer, Bhadra and Gitlin (1965) have reviewed previous work which demonstrates that these processes may also cause the nucleation of supercooled water, thus suggesting the operation of a cavitation mechanism. Experiments by Gitlin and Lin (1969) indicated that cavitation was a necessary but not sufficient condition for the dynamic nucleation of supercooled water.
They also explained the results of previous experiments by Goyer et al. (1965) on the shock-induced freezing of supercooled water in terms of cavitation in the samples of supercooled water containing thermocouples, the thermocouple providing an excellent site for cavity formation when the water is accelerated away from it by the shock wave. Cavitation may also be produced by the disruption of a drop when subjected to electrical or mechanical disruption since, at the point of maximum stress, the internal drop pressure must undergo a substantial reduction.

Cavities formed by the processes outlined above will collapse violently upon removal of the initial stress. This collapse involves an inrushing of liquid to fill up a near vacuum. Hickling and Plesset (1964) calculated the maximum pressures which may be expected from cavity collapse in water. Their values range from several tens to several hundreds of kilobars, depending upon the amount and thermal behaviour of any gas present in the cavity, the presence of gas generally giving rise to the higher instantaneous pressures.

Two mechanisms have been proposed in an attempt to explain the means by which the growth and subsequent collapse of a cavity might lead to the nucleation of solid in a supercooled liquid. The first, proposed by Chalmers (1963), is that freezing is induced by evaporative cooling at the surface of a growing bubble. However, Hickling (1963) showed that because of the rapid increase of pressure within the bubble, resulting from evaporation, the maximum cooling will be about 2°C. The second possibility, considered in more detail in the following paragraphs, is that nucleation may result from the change in equilibrium freezing temperature produced by the pressure changes which accompany the collapse of a cavity.

In a comprehensive review of the nucleation of freezing by cavity collapse, Hickling (1965) demonstrated that embryo ice crystals may be produced by the influence of the high pressures generated by this means upon the equilibrium melting temperature of ice. Fig. 6 presents the ice-water phase diagram and it may be seen that the equilibrium melting temperature of water is of the order of 150°C at pressures of a few tens of kilobars. The water will also be subject to a local temperature rise due to adiabatic compression and the dotted line shows how the temperature of the water rises due to this compression from an initial state at 1 atmosphere and 0°C. It can thus be seen that a sub-cooling of about 100°C at $4 \times 10^4$ atmospheres may be produced for water at an initial temperature and pressure of 0°C and 1 atmosphere respectively. This hypothesis is supported by evidence that liquids whose melting points increase continuously with pressure may be readily nucleated by mechanical disturbances believed to cause cavitation.

The behaviour of a cavitation bubble after the initial collapse and rebound is not known in detail. However, according to Hickling, if the water is originally in a supercooled state it seems reasonably certain that the ice particles that have been created can return to an environment of water at its original temperature without serious exposure to superheating. Thereafter, freezing is assumed to occur with these particles acting as nuclei. The crystalline structure of these particles would be different from that of ordinary ice but Hickling suggests that they are capable of acting as effective nuclei either by means of a preliminary solid-phase transformation or through heterogeneous nucleation. Chalmers (1964) has shown that the size of these particles is appropriate for nucleation in water at one or two degrees of supercooling. The evidence discussed by Hickling (1966) for a reduction in cavitation damage to metals immersed in water as the temperature is reduced below about 50°C provides corroborative support for the suggestion that ice can be nucleated in water by means of cavity collapse. Several experiments were performed in order to test this cavitation hypothesis.

In the first experiment, pairs of drops were mounted upon insulating rods and allowed to cool between two electrodes. When the drops had cooled to their environmental temperature, an electric field of sufficient magnitude to cause their disruption was applied between the electrodes and the process of disruption and subsequent freezing was recorded using a high-speed camera. One typical photographic sequence is reproduced in Fig. 7, from which it may be seen that the freezing is initiated from the interface between the drop and
its support. In all the cases observed, the freezing was definitely associated with the disruption of the supported drops and was never a consequence of a simple movement of the triple-phase interface. It is proposed that the sharp reduction of pressure within the drop upon disruption is sufficient to cause the formation of cavities which will form preferentially at the water-solid interface where favourable sites may be expected to occur.

The second experiment involved the passage of a spark discharge to the surface of supercooled water situated in a glass dish which contained a brass earth electrode upon which was mounted a thermocouple in order to measure the water temperature. A spark could be passed from a metallic needle situated just above the dish, to the water surface, the spark and any subsequent growth of ice being recorded by a 16 mm movie camera. It was found that, immediately after the passage of the spark, ice formed, in the great majority of cases, at the earth electrode and propagated from there into the surrounding water at velocities very close to those predicted by Hallett (1964). A photographic sequence showing the growth of ice following the passage of a spark is displayed in Fig. 8. These
Figure 7. Stages in the freezing of a pair of supercooled drops of radius 1.0 mm disrupted by an electric field at a temperature of $-10^\circ$C. A, time $t = 0$ s; B, $t = 0.003$ s; C, $t = 0.006$ s; D, $t = 0.011$ s; E, $t = 0.017$ s; F, $t = 0.020$ s; G, $t = 0.024$ s; H, $t = 0.028$ s; J, $t = 0.032$ s.
Figure 8. The growth of ice in bulk supercooled water of temperature $-3.0^\circ C$ after the passage of a spark to the water surface at a time $t = 0$. A, $t = 0$ s; B, $t = 0.06$ s; C, $t = 0.12$ s; D, $t = 0.18$ s; E, $t = 0.31$ s; F, $t = 0.63$ s; G, $t = 1.25$ s; H, $t = 1.88$ s; I, $t = 3.13$ s; J, $t = 5.0$ s.
Figure 9. Stages in the formation and collapse of a cavity within a warm water drop of diameter 3·0 mm disrupted by an electric field at a time \( t = 0 \). The sequence is down the first column, then the second and then the third, corresponding to times: \( t = 0·75; t = 1·00; 1·23; 1·75; 2·00; 2·25; 2·50; 2·75 \) millisecond.
Figure 10. Stages in the formation and collapse of a cavity and the growth of ice within a supercooled water drop of temperature $-12^\circ C$ and diameter 3.0 mm disrupted by an electric field at a time $t = 0$. The sequence is down the first column and then the second, corresponding to times: $t = 0.2; 0.4; 0.6; 0.8; 1.2; 1.2$ millisec.
observations may be explained in terms of the growth and collapse of cavities formed by the shock waves since the earth electrode would offer a particularly suitable site for cavity formation. Also, the observation that an electrode connected to earth was much more efficient than a similar but isolated electrode in causing nucleation suggests that a small amount of electrolysis might assist the development of cavities.

A further series of experiments was conducted in an attempt to establish photographically whether the disruption of a supercooled water drop was accompanied by the growth and collapse of a cavity, followed by freezing. The principal instrument utilized was a high-speed 16 mm camera capable of framing rates of up to 11,000 s⁻¹.

A water drop of diameter typically about 3·0 mm was suspended from the flat base of a highly polished cylindrical perspex rod of length 13 mm and diameter 3·0 mm. The rod passed snugly through a hole in the upper of a pair of brass electrodes across which a preselected high electric field could be applied. The drop on the base of the rod was thereby positioned in the centre of the electrode system. The interface between the drop and the base of the rod could be viewed by looking down the rod through a microscope of magnification such that the perimeter of the interface defined the field of view. The interface could be illuminated using an appropriate optical system and could be photographed through the microscope by means of the high-speed camera.

The experimental procedure was to mount the drop on the rod, to allow it to come into thermal equilibrium with its environment and then to activate an electronic timing arrangement by which the drop was disrupted just as the light-source achieved maximum intensity and the camera reached full speed. Experiments were performed at room temperature and, by locating the equipment inside a refrigerator, at sub-zero temperatures ranging from −8°C to −12°C. Pure water and water deliberately contaminated with sulphur dioxide were used.

Fig. 9 shows a sequence of high-speed photographs illustrating the growth, oscillation and collapse of a cavity when a drop of pure water was disrupted at a temperature of +20°C. Typical values for cavity diameter, duration and oscillation frequency are seen to be about 300 μm, 3 m s⁻¹ and 1,000 s⁻¹ respectively, which are in reasonable agreement with those determined experimentally and theoretically in much larger volumes of water by Noltingk and Neppiras (1950), Knapp and Hollander (1948), and Hickling (1966). Similar series of photographs were obtained with supercooled drops and with drops contaminated with sulphur dioxide. Photographs of undisrupted drops inoculated with small air bubbles were found to be very similar in appearance to those presented in Fig. 9.

Fig. 10 shows the growth and collapse of a cavity in a supercooled drop, followed by the development of the freezing process. A crescent-shaped pin was scored into the perspex base prior to the experiment in order to provide a favourable site for cavity formation. It is located on the upper left-hand side of the photographs about halfway between the centre and the edge of the drop and is visible on several of the frames. It is seen that the cavity was formed at this point. Freezing apparently occurred, according to the photographic evidence, at the edge of the drop but it is interesting to note that the measured time interval between cavity collapse and the first observation of the presence of ice agreed to within 5 per cent with that calculated for ice crystals to travel from the cavity to the edge according to the equation of Hallett (1964). This excellent agreement was confirmed in the other such experiment that was conducted. It suggests that nucleation occurred at the site of the cavity at the moment of collapse, but that the growing crystals could not be discerned until they started to propagate back into the drop from the edge.

The cumulative evidence from the experiments described in the foregoing paragraphs suggests strongly that cavitation is the mechanism by which freezing is initiated when supercooled drops are catastrophically disrupted.

4. Discussion

Nucleation of a supercooled liquid by a mechanism associated with the high pressures produced locally by the collapse of a cavity seems likely in a number of situations in which
cavities may be created. It has not yet been demonstrated, however, that cavitation may be produced in typical atmospheric situations although it seems reasonable to suppose that cavities may be formed when the surface of a drop is disrupted by either mechanical or electrical means. The formation of cavities is not always a sufficient condition for nucleation but there are indications that smaller ultrasonic or shock wave intensities are required for nucleation as the degree of supercooling is increased. It appears, therefore, that the important factor in this process is the increased effective supercooling produced by the high pressure pulse associated with the cavity collapse. Thus cavitation cannot be effective unless the cavity reaches a critical size, related to the degree of supercooling, before collapsing. The growth of larger cavities may be facilitated by the diffusion of dissolved gas into the growing cavity but it is not clear what the overall effect of the presence of this gas would be upon the nucleation process.

Evaporative cooling from the tip of a filament produced on disruption of a drop is unlikely to be sufficient to cause nucleation of the supercooled drop in all but the most extreme laboratory situations and will not be of significance in the atmosphere. The formation of minute crystallites by the process suggested by Loebl could be of importance in natural clouds but it is as yet supported by very little evidence.

The freezing of supercooled drops by disruption, whether mechanical or electrical, is possibly relevant to two distinct atmospheric situations. In the later stages of thunderstorm development, the electric field within regions of a thunderstorm will be sufficiently intense to disrupt the larger supercooled drops present and thus bring about their nucleation. Such a process could give rise to a 'hail gush' as mentioned earlier. Secondly, interactions between droplets in supercooled clouds may cause freezing if a filament is drawn out between them as a result of glancing collisions or the magnified electric field strength between the drops. Experiments involving falling droplet-pairs are required, however, before a conclusive assessment of this possibility can be made.

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