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THE ELECTRIFICATION OF WATER DROPS ON FREEZING OR MELTING AT TERMINAL VELOCITY IN AIR.

by

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THE ELECTRIFICATION OF WATER DROPS ON FREEZING OR MELTING AT TERMINAL VELOCITY IN AIR.

ABSTRACT.

Measurements on the electrification on freezing of individual water drops of diameter between 3 and 5 mm supported by an air stream have shown that the freezing behaviour of the drops is temperature-dependent. Above -10° C the freezing progressed uniformly throughout the drop from a single point. Below -10° C the outer surface of the drop froze rapidly with the exception of a small area at the top of the drop. Freezing then progressed uniformly from the base of the drop upwards. No shattering or electrification of the drops was observed during their freezing. These results are contrasted with those of previous workers who observed the freezing of drops suspended on fibres.

Measurements of the electrification of ice spheres supported by an air stream have indicated that the sign of the charge acquired by the spheres due to riming is temperature-dependent. When the sphere is rimed by droplets at temperatures above -10° C it acquires a negative charge, and when the riming droplets are at temperatures below -10° C the ice sphere acquires a positive charge. An explanation is proposed for this effect in terms of the observed freezing behaviour of individual water drops. It is suggested that this effect could lead to thunderstorm electrification of the observed polarity.

Measurements on the electrification of melting ice spheres supported by an air stream indicated that the sign of the charge acquired by the spheres is dependent on whether or not water is flung off from the sphere during melting. If no water was flung off the charge was positive, while if water was flung off the charge was negative. This may explain the discrepancies between the laboratory and field measurements of MacCready and Proudfit.

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CHAPTER 1.

THE THUNDERSTORM.

1. INTRODUCTION.

In attempting to portray the various aspects of a thunderstorm one is necessarily cautious in presenting a picture of a typical thunderstorm. This is because of the variation of the characteristics of thunderstorms over a wide range. In attempting to explain the electrification of a thunderstorm the theorist should seek a mechanism which will cover the whole range of storms.

2. METEOROLOGICAL AND PHYSICAL ASPECTS OF A TYPICAL THUNDERSTORM.

The most intensive study of cumulonimbus clouds which developed into thunderstorms has been described by Byers and Braham (1949). Radar and aircraft measurements showed a cumulonimbus cloud to consist of one or more identifiable units or cells, which were local regions of pronounced vertical velocity, precipitation and electrical activity. These cells range in size from 0.5 km to 10 km in horizontal extent. Küttner (1950) has suggested the existence of sub-cells of about 100 m horizontal diameter. Each cell has a life cycle which may be divided into three stages, these being growth, maturity and decay. During the mature stage of the cell, which lasts from 15 to 20 minutes, there is a strong central updraught, typically of 10 m sec^{-1} but sometimes as high as 50 m sec^{-1}, which may display a pulse-like nature (Workman and Holzer 1942). The columnar updraught is surrounded by a strong downdraught. Intense precipitation and electrical activity occur during the mature stage. Hail falls from the regions of the cell where the updraught is too weak to support it and may reach the ground either as hail or rain. In tropical thunderstorms very large hailstones may be formed by continued recirculation through the cell; elsewhere, however, the usual form is as small hail and soft hail with rounded or conical shapes and from 4 to 6 mm across. Precipitation as hail is usually intense and may reach 5 cm hr^{-1} (Latham & Mason 1961). During the later stages of the mature period the hail may give way to snow.

Also characteristic of a typical thunderstorm are its great vertical depth and the very wide temperature range spanned. The bases are usually warmer than 0° C and the tops usually extend beyond the -34° C. isotherm (Jones 1950) and may occasionally extend into the tropopause. The cloud top is characterised by an anvil-shaped mass of ice crystals which has the diffuse outline of cirrus cloud.

Lightning activity has been reported from clouds which have a different physical appearance from the above picture and whose tops were warmer than 0° C (Moore 1965). Although detailed observations of this type of activity are lacking the many reported cases suggest that it is a real effect.

The particular sizes and concentrations of the cloud particles found in a thundercloud are characteristic and they have been involved in various theories proposed to explain the electrification of the cloud. The size spectra of ice crystals in thunderclouds was measured by Jones (1960) who found the most common to be between 80 and 175μ in diameter and present in concentrations of 10^5 m^{-3}. Larger crystals were observed up to 5 mm., but the concentration fell off rapidly with increasing size and the larger crystals were only found near the 0° C isotherm. The droplet size spectra is less well documented, probably because of the greater difficulties involved in sampling, but the larger drop sizes are more numerous than in other types of cloud. Latham and Mason (1961b) quote the Meteorological Research Flight findings that droplets of diameter greater than 50μ are present in concentrations of at least 10^6 m^{-3}.

3. ELECTRICAL ASPECTS OF A TYPICAL THUNDERSTORM.

Electrostatic measurements at the ground do not give a unique picture of the distribution of charge in a thundercloud and so the distribution of charge is difficult to determine. However, the results of different workers using different techniques (see Chalmers (1956), Mason (1957)) have shown that the electric charge in a typical thunderstorm may be represented as a simple dipole with the positive charge uppermost. Often a region of smaller positive charge exists beneath the main negative charge and is usually below the 0^o C isotherm. It is not yet known whether this small charge is a feature of all thunderstorms. The magnitude of the charges neutralised by lightning discharges have been calculated from the change in electric moment caused by the discharge. The change in electric moment was deduced from potential gradient measurements at the ground. Pierce (1955) gives a value of 100 C km for the average electric moment change.

due to a discharge. For a charge separation of 5 km this gives the magnitude of the segregated charges as 20 C. In order to explain the charging current in the cloud in terms of gravitational separation of unsegregated charge it can be shown that approximately 1000 C of unsegregated positive and negative charges must exist in the region between the segregated charges. The expression relating the charge being separated after a discharge to the relative velocity V of the charge carriers has been calculated as 8000/V C.

When a lightning discharge occurs the potential gradient inside the cloud falls to zero and then immediately begins to recover in an exponential manner with a time constant of approximately 7 seconds. Wormell (1953) showed that this was consistent with the picture of a constant charging current and a dissipating current which is proportional to the charge separated. It is the constant current which theories of charge separation attempt to account for. Wormell found the value of this current to be 3A for a typical storm. The wide variation in the parameters of thunderstorms may be seen from the observations of Vonnegut and Moore (1958). They observed storms with from 10 to 20 discharges per second which means that the charging current must have been of the order of 100A.

In addition to the internal currents present in a thundercloud there are also external currents. There is a flow of positive ions to the base of the cloud due to point discharge at the ground. Chalmers (1953) has calculated this to be less than 0.5A. There is also a flow of current which carries positive charge from the top of the cloud to the ionosphere. Gish and Wait (1950) found this current to have an average value of 0.5A while Stergis, Rein and Kangas (1957) measured it as 1.3A.

It has been stated above that the electrical activity of a thunderstorm cell occurs at the same stage of its life-cycle as the intense convection and the growth of precipitation particles large enough to give radar returns. It is of great importance that the relationship with time of these phenomena be more precisely expressed. The early radar work of Workman and Reynolds (1949) indicated that the electrical activity did not occur until several minutes after the first precipitation echo was observed. This, in turn indicated that the electrical activity was dependent on the formation of precipitation. However, Moore (1965) has observed the appearance of electrical activity before that of detectable precipitation and this result throws doubt on the temporal relationship of the growth of hail and the onset of electrical activity. However, this issue which is of importance in the formulation of any theory of charge separation will not be settled conclusively without many more observations.

4. THEORIES OF THE ELECTRIFICATION OF THUNDERSTORMS.

The following list gives the names of some of those who have suggested possible mechanisms of cloud and thundercloud electrification during this century.

Gerdien	1905	Rossman	1948
Simpson	1909	Workman & Reynolds	1948
Elster & Geitel	1913	Wall	1948
Wilson	1929	Vonnegut	1955
Gunn	1935	Wilson	1956
Simpson & Scrase	1937	Reynolds Brooke & Gourley	1957
Findeisen	1940	Latham & Mason	1961
Frenkel	1944	Sartor	1961
Dinger & Gunn	1946	Magono & Takahashi	1963
Grenet	1947	Reiter	1965

The eventual success of any theory of thunderstorm electrification depends on its being applicable to all aspects of all thunderstorms. Many early theories have been abandoned because they were based on ideas which subsequent experimentation has shown to be incorrect, or to give insufficient electrification. However, many of the suggested mechanisms have not as yet been fully evaluated including some of the earliest theories. It is clear that several of these mechanisms will give rise to electrification in clouds but it is not certain whether any one mechanism will suffice or whether the electrification is due to a variety of mechanisms acting in concert.

Charge production mechanisms in clouds may be separated into three classes depending upon the origin of the positive and negative charges:

(1) Classical, in which charge is generated by the interaction of initially neutral cloud particles.

(2) Influence, in which positive and negative charges are already available as atmospheric ions.

(3) Convection, in which the origin of the charge is outside the cloud.

A combination of these processes is also possible. The charge may be separated by electrical or gravitational forces or, as suggested by Chalmers (1965), by a centrifugal force. Most current theories favour classical generation and gravitational separation in which charges are generated by the interaction of cloud particles of different sizes which are then separated because of the different fall-speeds of the particles. Some of the theories which have aroused considerable interest and controversy in recent years are described below.

(a) Simpson's Theory

Simpson (1909) found that the breaking of a water drop in a strong vertical air jet produced electrification of the resulting drops. The electrification was found to be sensitive to the presence of impurities in the water, but, for distilled water, the large fragments of the broken drops carried positive charges, while the surrounding air acquired a net negative charge. The charge produced was found to be of the order of 20×10^{-3} e.s.u. cm^{-3}, and to be independent of any initial charge on the drop before breaking. Simpson argued that the shattering of water drops in a thundercloud would give rise to large positively charged drops and a corresponding negative charge in the air. The negative charge would then be acquired by small cloud droplets and the charge would then be separated gravitationally. However, this would give rise to a charge distribution of opposite polarity to that observed.

Further, Mason (1957) calculates that the rate of building up of charge by this process is two orders of magnitude smaller than the required value.

(b) Wilson's Theory (1929)

In his 1929 theory Wilson suggested that the weak atmospheric electric field was capable of influencing the sign and growth of the electric field in a thundercloud. It is an influence theory and, like other influence theories, has fallen into disregard as it is unable to account for the required rate of charge separation in a thundercloud in the light of the available data. The principle of the theory is that water drops falling in the atmospheric electric field will be polarised by the field so that if their lower surfaces have a positive charge they will selectively capture negative ions from any cloud of ions or charged droplets through which they fall. These drops will fall to the bottom of the cloud thus augmenting the existing electric field. This will lead to further falling drops acquiring a greater negative charge and thus the electric field will be built up in this manner. The theory of the ion capture process has been worked out by Whipple and Chalmers (1944), and Chalmers (1947) has shown that a similar process applies to falling ice particles.

There are, however, limitations to the theory. In order to acquire a net negative charge the drops must be moving downwards faster than the downward moving positive ions. This is no longer true for small ions in electric fields of over 50,000 Vm^{-1} although selective capture of large ions and charged cloud droplets is possible in all fields less than the breakdown field. Wormell (1953) pointed out that the maximum possible rate of charging by this process cannot exceed the rate at which ions are produced in the lower atmosphere. This sets the maximum rate of charging at 6 C km^{-3} hr^{-1} as compared with the rate of 50C km^{-3} hr^{-1} considered to be necessary by Mason (1953). Further , Mason (1953) points out that unless the conductivity of the air in a thundercloud is unusually high the drops would only acquire a fraction of their maximum possible charge while in the thundercloud. There has apparently been no measurement of the conductivity inside a thundercloud as yet. If such measurements showed the conductivity to be unusually high then the source of the charges would have to be some process such as point discharge. Current opinion however is that the conductivity is not sufficiently high for the Wilson process to be considered as the major source of electrification although it may well be of importance in determining the charge carried by precipitation which has fallen through the region of dense space charge below the cloud.

(c) The Dinger-Gunn Effect.

As described in chapter 6 the experiments of Dinger and Gunn have shown that when samples of ice containing air bubbles melt in an airflow charge generation occurs so that the melt water becomes positively charged, and an equal and opposite charge is carried away by the air. Dinger and Gunn found the quantity of charge separated to be 1.25 e.s.u. for each gram of ice melted. They suggested that this mechanism might be of importance in the electrification of thunderclouds in regions where hailstones are melting. As regards the main dipole charge of the cloud this mechanism is unsatisfactory as it would produce charges of the opposite sign to that observed. Further, while it is difficult to estimate the value of a charging current due to melting hailstones it may be calculated that the maximum possible charge produced in this way in a cloud containing 2 g m^{-3} of solid precipitation elements would be 0.8 C km^{-3}, much less than the required concentration of 20 C km^{-3}. However, it is possible that the mechanism contributes to the lower positive charge of the thundercloud.

(d) The Workman - Reynolds Effect.

The setting-up of large potential differences across the ice-water boundary of freezing dilute aqueous solutions was discovered by Costa Ribeiro in 1945. This effect has been extensively investigated by Workman and Reynolds(1948, 1950). They measured the freezing potentials developed by water and dilute aqueous solutions, and found that potential differences of up to several hundreds of volts could be obtained, the magnitude and sign of the potential difference depending on the impurity in solution and its concentration. Measurements on samples of carefully purified water showed that only small potential differences were set up. It is not certain whether or not a freezing potential exists for absolutely pure water. The mechanism of this effect will be considered further in chapter 2. Workman and Reynolds have applied their results to a theory of thunderstorm electrification in which the wet hailstone is the charge generator. They argue that if a graupel particle falls through a cloud of supercooled water droplets and collects them at a sufficient rate to maintain a liquid film on their surface then only a small portion of any droplets collected will be frozen, the remainder being flung off from the top of the hailstone in the form of small drops. They suggest that the contaminants present will be such that the ice becomes negatively charged and that the droplets flung off will carry away a positive charge. The hailstone is thus left with a net negative charge. As the hailstone falls further through the cloud it will eventually start melting. At this point in the hailstone's life, the water drops which impinge on it will share the electric charge of the hailstone and eventually will reduce its charge to zero. The main advantages claimed for this theory by the authors are that it explains the centre of negative charge being at the -10° C isotherm, and that the quantities of charge separated are more than ample, up to 9×10^{4} e.s.u. cm^{-3} of water frozen for a solution of sodium chloride.

There have been a number of objections to this theory. Reynolds (1954) has pointed out that it is probable that glazing of hail, that is the formation of a liquid film on the hail, probably does not occur in the thundercloud at temperatures below -15° C, whereas charge separation certainly occurs at lower temperatures than this. Further, Reynolds found that an ice surface showed charging only in the presence of water drops and ice crystals. Also no charge separation has been observed on the glazing of an ice-coated probe by Reynolds et al (1957), Latham and Mason (1961), Magono and Takahashi (1963) or Church (1966). Apart from these results the charge generation mechanism is sensitive to the type and quantity of impurity present, and for it to be successful in producing the electrification of a thunderstorm it would seem to require an ideal concentration of impurities.

(e) Convection Theories.

Grenet (1947) suggested that a convection process was responsible for cloud electrification and this was supported in greater detail by Vonnegut (1955). Vonnegut put forward several arguments against precipitation being the principal charge generating mechanism. The major ones are that there is no relationship between rate of precipitation and intensity of electrification, that the charge measured on precipitation from thunderclouds is very small and usually of the wrong sign, and that the violent updraught would interfere with any orderly charge separation. In Vonnegut's theory the charges are transported in the updraughts and downdraughts of the storm. He suggests a process in which positive space charge from the lower regions of the atmosphere is carried by the updraught to the top of the cloud where it attracts negative ions from the ionosphere. These negative ions are captured by cloud particles and move downwards in the downdraught. At the base of the cloud this negative charge attracts more positive space charge and, if the potential gradient becomes sufficiently large, ions from point discharge at the ground. These positive charges are then carried to the top of the cloud and the process continues until large potential gradients are built up.

Wilson (1956) proposed a theory which was a combination of his earlier influence theory and a convection theory. He suggests that the process suggested in his 1929 theory first builds up the charge separation in the cloud. Positive ions from point discharge then attach themselves to cloud droplets which rise to the top of the cloud and attract negative ions down from the ionosphere. These negative ions attach themselves to precipitation particles and fall to the base of the cloud to form the main negative charge.

The outstanding difficulty of both these theories is the question of why the ions of the right sign attach themselves preferentially to one type of cloud particle. rather than to the other. However, a final decision on both of these theories will have to await the results of further investigations into the movement of air masses in thunderclouds.

(f) The Theory of Reynolds, Brook and Gourley.

From a laboratory investigation into the electrification due to the collisions of ice crystals with simulated hailstones in the presence of cloud droplets, Reynolds, Brook and Gourley (1957) estimated that the mean charge separated by a rebounding ice crystal was 5×10^{-4} e.s.u., leaving the hailstone negatively charged. They applied this result to a model thundercloud containing a crystal concentration of 10^4 m^{-3} and a liquid water concentration of 1 gm m^{-3} and showed that hail in concentrations of 10 g m^{-3} falling at 10 m sec^{-1} relative to the ice crystals was capable of producing a discharge of 20 C in a thundercloud cell of 1 km diameter in about 14 minutes. In a thundercloud of volume 50 km^3 the process would separate sufficient charge to account for the observed rate of repetition of lightning discharges.

The experimental techniques used in this investigation appear to be subject to considerable error, and confirmation is desirable. Latham and Mason (1961) performed further experiments of this type and found the charge separated to be five orders of magnitude less than that found by Reynolds et al., while Church (1966) found it to be two orders of magnitude less. This spread in the observed values of the charge separation means that before the charge produced by the collision of ice crystals and hailstones can be evaluated as a prime source of charge in a thundercloud further experimentation is necessary.

(g) <u>The Latham-Mason Theory</u>.

Latham and Mason (1961) estimated the charge separated by the shattering of supercooled water drops freezing onto hailstones and concluded that the electrification produced by the shattering of droplets in the range $40 - 100\mu$ would be sufficient to separate charge at the rate of 1 C km^{-3} min^{-1}. This is sufficient for the average thunderstorm but does not account for the separation of charge in violent thunderstorms which may be up to 100 times greater. This difficulty cannot be easily resolved by scaling up either the droplet or hailstone concentrations. This theory will be considered in more detail in chapter 2.

(h) <u>Sartor's Theory</u>.

Sartor (1961) has proposed an inductive theory of electrification. He considers that particles in a cloud are polarised by the electric field and that when they collide without coalescence, or pass within a short distance of each other, charge is generated and then separated by the action of gravity. This theory is a more general form of the theories of Elster and Geitel (1913) who considered the charge separation between polarised water droplets which collided without coalescence, and of Müller-Hillebrand (1954) who considered the charge separation between ice crystals and hailstones polarised in an electric field. It is also similar to Wilson's influence theory (1929). Sartor showed that the larger polarised cloud particles would become negatively charged, and that if they grew by absorbing smaller charged cloud particles both their charge and their fall speed would be enhanced, thus causing a rapid building up of the field. He considered that ice particles would be more important than water drops as their greater rate of growth would outweigh their smaller fall speeds. He also showed that this mechanism would cause the electric field to increase exponentially and calculated the rate of charge separation for various droplet separation efficiencies. The droplet separation efficiency is the fraction of droplets which do not coalesce after colliding. He found that for a separation efficiency of 0.1 the electric field in a thundercloud would increase by a factor of 1000 in 10 minutes. Recent measurements on the charge separation between model ice crystals which do not touch (Latham, Mystrom and Sartor, 1965) have shown that the charge separation between ice particles is probably greater than that between water drops as appreciable quantities of charge can be separated between ice crystals separated by distances of the order of their dimensions.

Sartor's theory is a powerful one but more information concerning the interaction of ice crystals and water drops in electric fields is required before its application to thunderstorm electrification can be more than speculation.

(i) Reiter's Theory

Reiter (1965) has discovered a mechanism which, it is claimed, will separate charge in a thundercloud at the rate of 15 C km^{-3} min^{-1}. Although this is 15 times greater than the value considered to be sufficient to produce the main charge by Latham and Mason (1961) he puts it forward as a subsidiary charge separation mechanism. Reiter has shown that appreciable quantities of nitrate ions are produced in clouds, mainly by silent electrical discharges, and that the greater the degree of atmospheric instability the greater the numbers of ions produced. Laboratory experiments showed that when ice particles grown by sublimation broke away from a cold plate they carried away 10 - 50 times as much charge when they were allowed to grow in an atmosphere containing nitrous gases than when they were grown in an atmosphere of ordinary air. Reiter applied these results to thunderclouds, and suggested that some form of atmospheric feedback occurred in which the charge separation mechanism was the fragmentation of crystal dendrites and needles and splinters from the surface of hailstones in a nitrous atmosphere. It was suggested that these charged particles would somehow be segregated in a way which would increase the field and also the nitrate ion concentration, thus increasing the charge separated by the fragmentation processes.

This theory gives rise to a number of questions. Although it explains why the electrification is more intense in a thundercloud than in other clouds because of the greater degree of atmospheric instability associated with thunderclouds, it does not explain why, once the charge separation has commenced in any small cloud, the electrification does not build up to thundercloud levels. It does not explain how the charged particles are separated so as to enhance the field and no evidence is given as to the amount of ice splintering which occurs in thunderclouds. There is also the question of how valid the laboratory results are when applied to the conditions in a thundercloud. These issues merit further investigation.

CHAPTER 2

SOME THEORIES OF THE ELECTRIFICATION OF ICE AND WATER.

1. INTRODUCTION.

As can be seen from chapter 1 many of the proposed theories of charge generation in thunderclouds depend on the electrification of ice and water. In this chapter the electrical properties of ice are considered in terms of its physical structure, and comparisons are made with the behaviour of water.

2. THE STRUCTURE OF ICE AND WATER.

(a) The structure of ice.

Apart from some high-pressure forms, about which little is known, only three ice structures are known to exist:

(1) the normal hexagonal structure.
(2) a cubic structure.
(3) an amorphous structure.

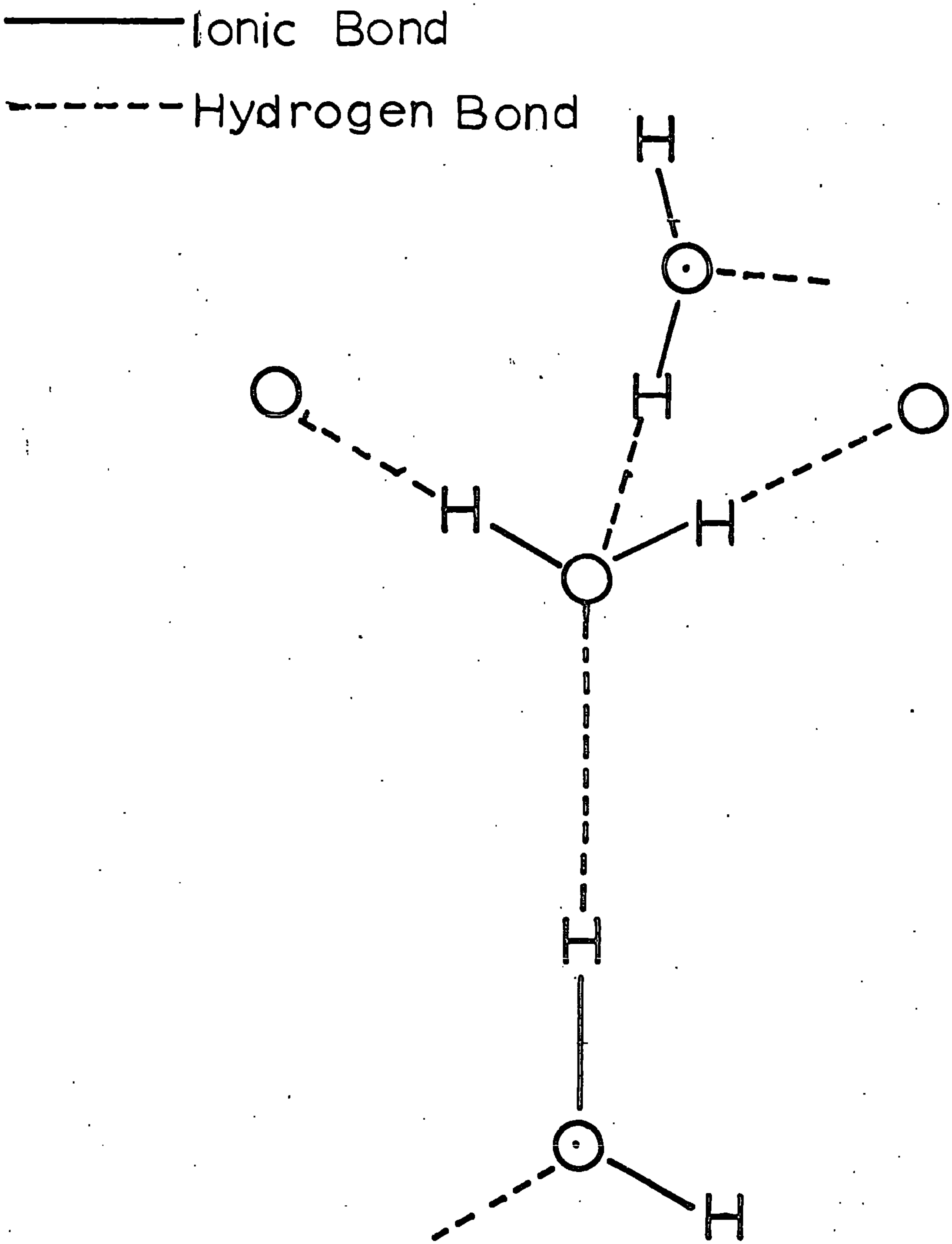
At normal pressures the hexagonal form is stable over the temperature range encountered in the atmosphere, and is the only form which is found in atmospheric ice particles. The structure of ice was first analysed by Barnes (1929). Bernal and Fowler (1933) discovered the importance of the hydrogen bond in determining the structure of ice and put forward three suggestions as to the nature of the hydrogen bonding:

(1) the hydrogen atoms be on the lines connecting neighbouring oxygen atoms;

(2) there is only one hydrogen atom on each such linkage thus forming hydrogen bonds;

(3) each oxygen atom has two hydrogen atoms at a short distance and hence water molecules are preserved.

These three statements are usually referred to as the Bernal-Fowler rules. Pauling (1935) pointed out that an ordered arrangement of hydrogen atoms according to the Bernal-Fowler rules would be inconsistent with the finite zero-point entropy observed by Giauque and Ashley (1933). He explained this by considering that the hydrogen atoms were able to move between different equilibrium positions on the lines joining the oxygen atoms. This model is well supported by X-ray and neutron diffraction studies. The oxygen atoms have been found to be separated by 2.76Å, and the hydrogen atoms to be on the lines joining the oxygen atoms and at a distance of 1Å from them. The hydrogen atoms have been found to alternate between the two possible equilibrium positions on each bond, and to spend half of the time in each. This is illustrated in Fig.1.



O—H Bond Length = 1·00Å

O----H Bond Length = 1·76Å

Fig.1 The Structure of Ice.

(b) The structure of water.

The structure of water is still the subject of debate, Bernal and Fowler (1933) suggested that water has a tetrahedrally hydrogen-bonded structure between 0^{o} C and 200^{o} C, and that between 4^{o} C and 200^{o} C the lattice structure is similar to that of quartz, while below 4^{o} C the structure is similar to that of the tridymite form of silica. Pauling proposed a pentagonal dodecahedron structure in which hydrogen bonding is of prime importance, and van Panthaleon van Eck et al (1958) have suggested a tetrahedral structure, similar to that of tin, in which the bonding is of the Van der Waals' type. The degree of order in water is determined by a statistical process which is dependent on temperature, increasing with decreasing temperature.

3. CHARGE TRANSPORT IN ICE AND WATER.

(a) Charge transport in ice.

Measurements on the electrical conductivity of ice have shown it to be slightly greater than $10^{-9}\,\Omega^{-1}\,cm^{-1}$ (Gränicher et al, 1957). Decroly et al (1957) have shown the static conductivity to be purely ionic. Gränicher (1958) points out that the ionic conductivity can be explained if ionised states exist in the lattice, the ions being the hydroxonium ion, H_3O^+, and the hydroxyl ion, OH^-. The manner in which an H_3O^+ ion diffuses through the lattice is illustrated in Fig. 2. The generation and diffusion of ions proceed by the translational motion of protons from one molecule to its neighbour. Reference to Fig. 2 shows that the diffusion of an H_3O^+ ion leads to a re-orientation of molecule β so that further diffusion of ion-states is not possible. The OH^- ions diffuse in a similar manner. The existence of ion-states alone cannot account for the observed time-independence of the conductivity, as the re-orientation caused by ion diffusion would tend to decrease the conductivity with time. In order to explain the time-independence of the conductivity it is necessary to postulate the existence of lattice defects whose diffusion through the lattice re-orientates the molecules so as to allow the further diffusion of ion-states. These are the Bjerrum defects which are generated by rotational movement of a proton in its own molecule, thus violating the third Bernal-Fowler rule. Each such jump generates one doubly occupied bond, or D- defect, and one vacant bond, or L- defect. The manner in which D- defects diffuse through the lattice is shown in Fig. 3. The L- defects diffuse in a similar manner. Reference to Fig. 3 shows that the diffusion of D- defects re-orientates molecule β so as to allow further diffusion of ion-states. As the re-orientation occurs in

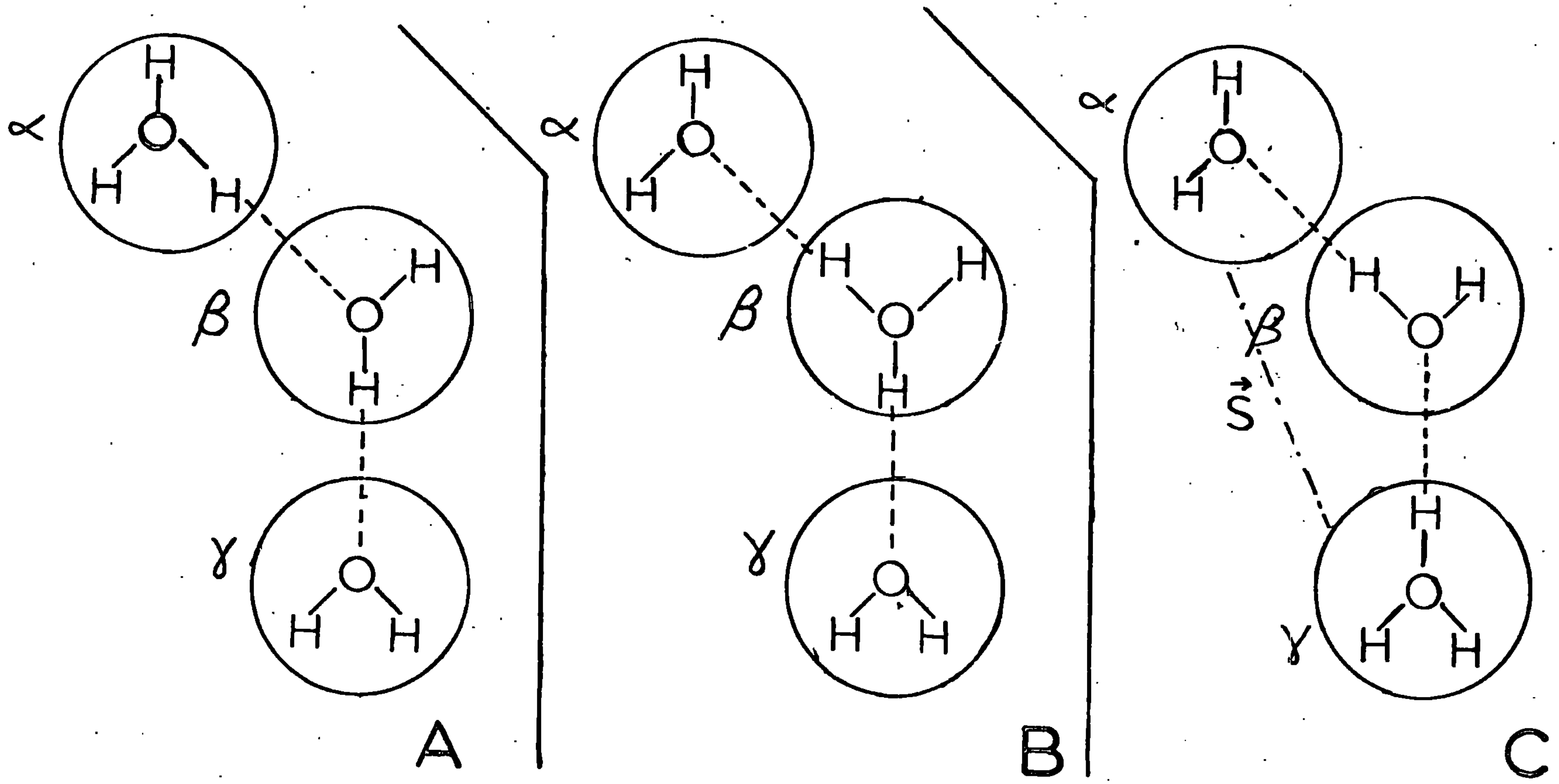


Fig. 2 Successive Steps in the Diffusion of a H_3O^+ Ion;
$\vec{S}$ = Ion Displacement.

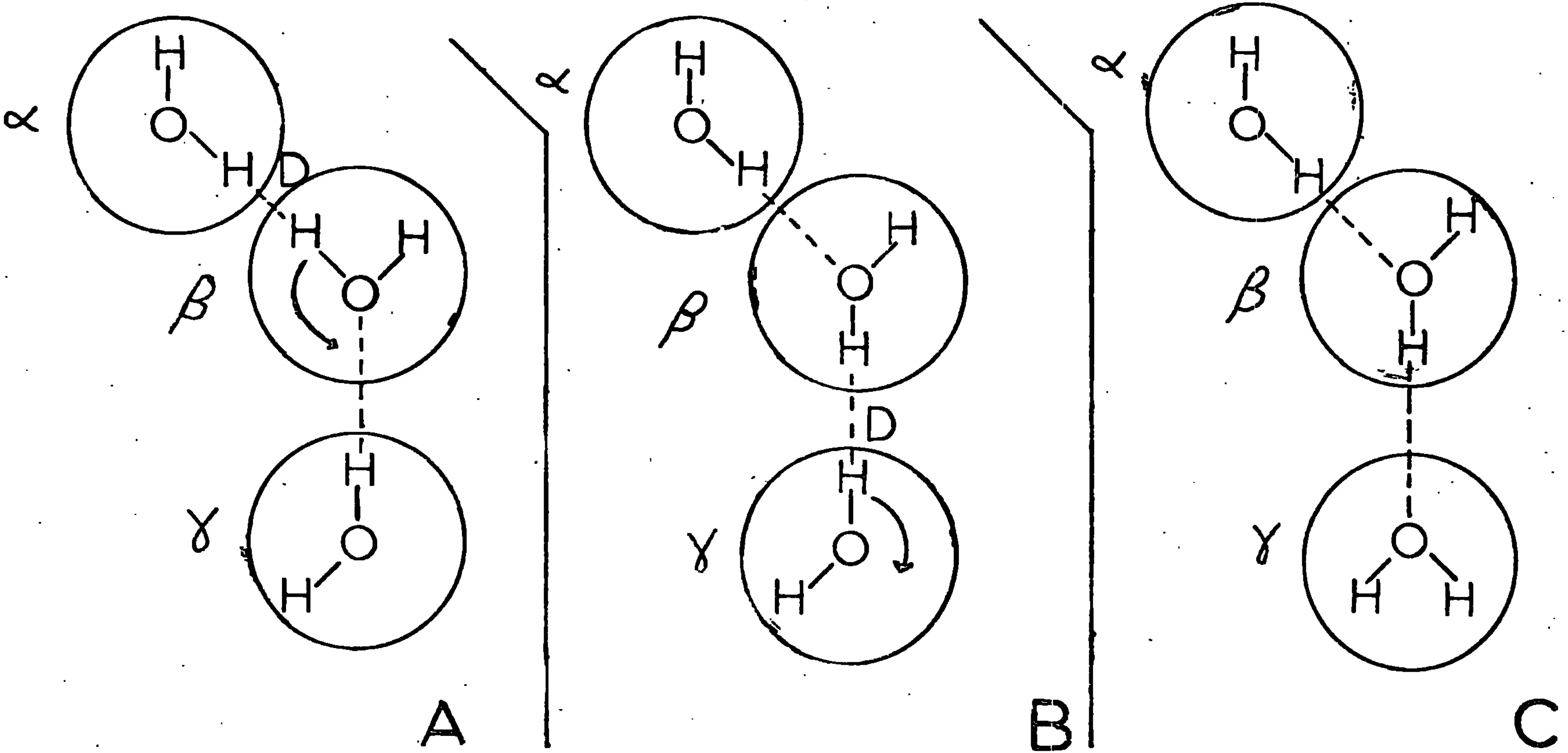


Fig. 3. Successive Steps in the Diffusion of a D-defect.

a time which is short compared with the arrival of protons at molecule α , the ion state diffusion process is governed by the rate of tunnelling of protons along the hydrogen bonds. This is high, thus explaining why the mobility of protons is higher in ice than in water.

Table 1 gives comparative figures for the properties of ion-states and Bjerrum defects. It may be seen that the Bjerrum defects are more numerous, but that their mobility ratio is approximately unity. Exact mobility ratios have not been measured for either ion-states or Bjerrum defects. The value for the ion-states is that found by Eigen and de Maeyer (1958).

<u>Table 1. Comparative values of the properties of ion-states and Bjerrum defects in pure ice at -10° C.</u>

Property	Ion-states	Bjerrum defects.
Energy of formation	1.2 ± 0.1 eV	0.68 ± 0.04 eV
Concentration of defects	8×10^{10} cm^{-3}	7×10^{15} cm^{-3}
Transition probability	6×10^{13} sec^{-1}	2×10^{11} sec^{-1}
Mobility	$\mu^{+} = 7.5 \times 10^{-2}$	$\mu^{L} = 2 \times 10^{-4}$
	cm sec^{-1} per V cm-1	cm sec^{-1} per V cm^{-1}
Mobility ratio	$\mu^{+}/\mu^{-} = 10$ to 100	$\mu^{L}/\mu^{0} \gtrsim 1$
Activation energy of diffusion	0 (tunnelling)	0.235 ± 0.01 eV.

(b) Charge transport in water.

Charge transport in water is by means of the diffusion of the H_3O^+ and OH^- ions. The mobilities of these ions have been found experimentally at 25° C as follows

H_3O^+	36.2×10^{-4}	$cm\ sec^{-1}$ per Vcm^{-1}
OH^-	19.8×10^{-4}	$cm\ sec^{-1}$ per $V\ cm^{-1}$

These values are high as compared with other univalent ions in water, the mobilities of which range from 4.0×10^{-4} $cm\ sec^{-1}$ per $V\ cm^{-1}$ for Li^+ to 7.9×10^{-4} $cm\ sec^{-1}$ per $V\ cm^{-1}$ for Cl^- , and this suggests the existence of some special transport mechanism.

The theories which have been proposed over the last 30 years all assume the existence of proton jumping from molecule to molecule, as suggested by Hückel (1928). The most satisfactory theory to date is that of Gierer and Wirtz (1949), which predicts values of the ionic mobilities in good agreement with experiment, although later work has shown it not to be completely correct. Figure 4 shows how H_3O^+ and OH^- ion-states are transferred by proton jumping. Gierer and Wirtz proposed two alternative mechanisms by which the rate of ion-state diffusion is determined. Eigen and de Maeyer (1958) considered both mechanisms in the light of the experimental evidence,

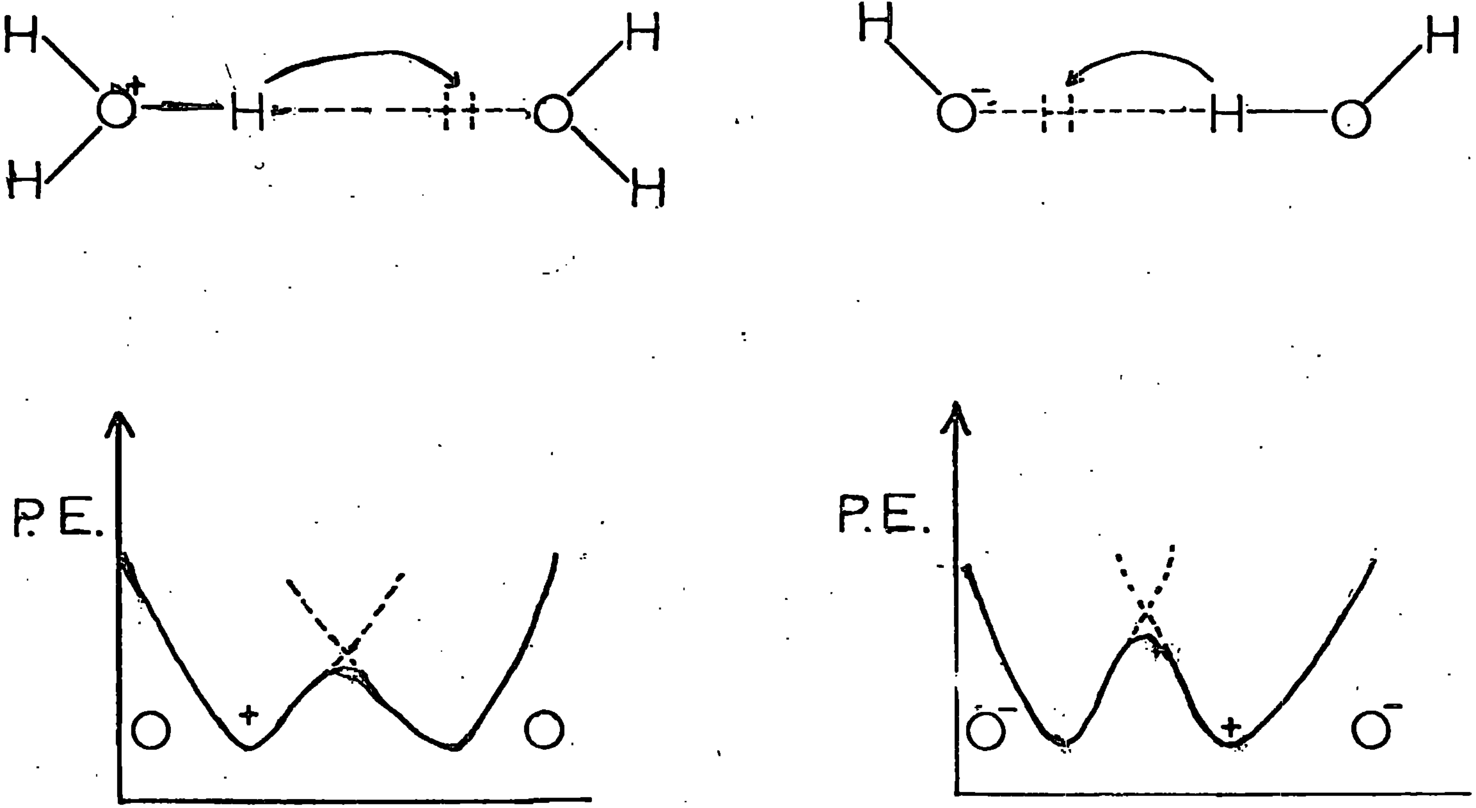


Fig.4. Transfer of "Excess" and "Defect" Proton in the Hydrogen Bond in Water.

and favoured the rate of formation of hydrogen bonds as being the determining factor.

4. ELECTRIFICATION PROCESSES IN ICE AND WATER.

(a) Charge separation in ice.

The various mechanisms for separating electric charge in solids have been considered in detail by Loeb (1958). Most of the mechanisms have no direct relevance to the study of ice as they depend on electrolytic effects and contact potential effects between dissimilar materials. The three mechanisms which seem most likely to lead to electrification in ice are piezoelectric and pyroelectric effects, and effects involving ionic conduction.

Piezoelectric effects are observed when externally applied stresses cause a material to become polarised. The most recent measurements of the piezoelectric effect in ice appear to be those of Teichmann and Schmidt (1965), who found the effect to be zero within the limits of their experimental accuracy. Pyroelectric effects have been found only in crystals which exhibit piezoelectricity and are detected as a charge separation produced by local heating. It is doubtful whether a true pyroelectric effect exists in the absence of stress. Mason and Owston (1952) attempted to detect both piezoelectric and pyroelectric effects in ice crystals grown from the vapour but were unable to do so. Mason (1957) has pointed out that the molecular structure of ice and the randomised motion of the dipoles militate against the lattices' having a resultant dipole moment.

Henry (1952) suggested a temperature gradient effect as an explanation of the charge separation resulting from the asymmetric rubbing of two pieces of identical insulating material. Under these conditions of contact the two pieces of material acquire equal and opposite quantities of charge. A formal theory of the temperature gradient effect in ice was put forward by Latham and Mason (1961) and this will now be considered in detail.

The temperature gradient theory for ice.

A charge separation mechanism due to the presence of temperature gradients in ice was first suggested by Findeisen (1940). A formal theory was proposed by Latham and Mason (1961 A). The mechanism is analogous to that of the Thomson effect in metal, except that the charge carriers are ions rather than free electrons. While it is unlikely that the positive charges are carried by free protons, but rather by H_3O^+ ions, the theory is developed in terms of the H^+ and OH^- ions. Essentially the mechanism depends on two facts:

(1) the concentrations of H^+ and OH^- ions in ice rise quite rapidly with temperature;

(2) the mobility of the H^+ ion is at least ten times greater than that of the OH^- ion.

Latham and Mason considered the charge separated under the influence of a steady temperature gradient, and also the charge separated by the transient contact of two pieces of ice at different temperatures.

(i) The separation of charge due to a uniform temperature gradient.

When a temperature gradient is set up in ice it will be accompanied by the setting up of a concentration gradient of ions of both signs. The more rapid initial diffusion of the H^+ ions down the concentration gradient will lead to a separation of charge with a net excess of positive charge at the colder end of the ice. The space charge set up in the ice due to this separation will produce an internal electric field which will tend to oppose further separation of charge. Latham and Mason point out that there might be a contribution due to the Bjerrum defects, but that as the ratio of their mobilities is approximately unity they have considered the charge separation to be due solely to the differential migration of ions. After some time a steady state is reached in which the effects of the concentration gradient and the internal electric field are balanced so that there is no net flow of current in the ice. Under these conditions, and assuming the charge separated to be concentrated on the ends of the ice, Latham and Mason were able to show that the predicted charge separation would be

$$\sigma = \frac{\mathcal{E} k}{8\pi e}\left(\frac{\mu^+/\mu^- - 1}{\mu^+/\mu^- + 1}\right)\left\{\frac{\phi}{kT} + 1\right\}\frac{dT}{dx}$$

where σ is the surface density of charge on the ends of the ice, $\mathcal{E}$ the static permittivity of ice, k Boltzmann's constant, e the electronic charge, μ^+/μ^- the mobility ratio for the H^+ and OH^- ions, ϕ the activation energy for dissociation in ice, T the absolute temperature, and dT/dx the temperature gradient. On substitution of values the charge density is given by

$$\sigma = 4.95 \times 10^{-5}\ (dT/dx)\ \text{e.s.u. cm}^{-2}$$

(ii) The separation of charge due to transient ice - ice contact.

When two pieces of ice, initially at different temperatures T_1, T_2, are brought into contact, a temperature gradient will be set up across the boundary plane ($x = 0$). After time t the temperature gradient will be given by

$$-\left(\frac{dT}{dx}\right)_{x=0} = \frac{(T_1 - T_2)}{2(\pi K T)^{\frac{1}{2}}}$$

where K is the thermal diffusivity of ice. From this equation it may be seen that the initial temperature gradient is very high, and so, therefore, is the net flow of current across the plane of contact. The rate at which charge separation proceeds is determined by the rate of diffusion of charge against the increasing internal electric field. The temperature gradient decreases rapidly and soon reaches the magnitude at which the flow of current due to it is equal and opposite to that due to the internal field. At this time the charge separation is at a maximum, and then decreases with time. Using a similar argument to that for uniform temperature gradients, Latham and Mason were able to deduce the variation of charge separated with time, and to show that the maximum separation occurs after a contact time of 8.5×10^{-3} seconds, and has the value

$$\underline{\sigma = 3.05 \times 10^{-3} \ (T_1 - T_2) \ \text{e.s.u.} \ \text{cm}^{-2}}$$

The variation with time of the charge separated is shown in Fig. 5.

Latham and Mason investigated the charge separation experimentally and found good agreement with the theoretically predicted values, except in cases where part of the ice was warmer than - 7° C. They related this with the variation of the electrical conductivity of ice with temperature as found by Bradley (1957). When they allowed for this variation in the theory, the experimental and theoretical results agreed closely. They also obtained evidence that the development of proportionately larger

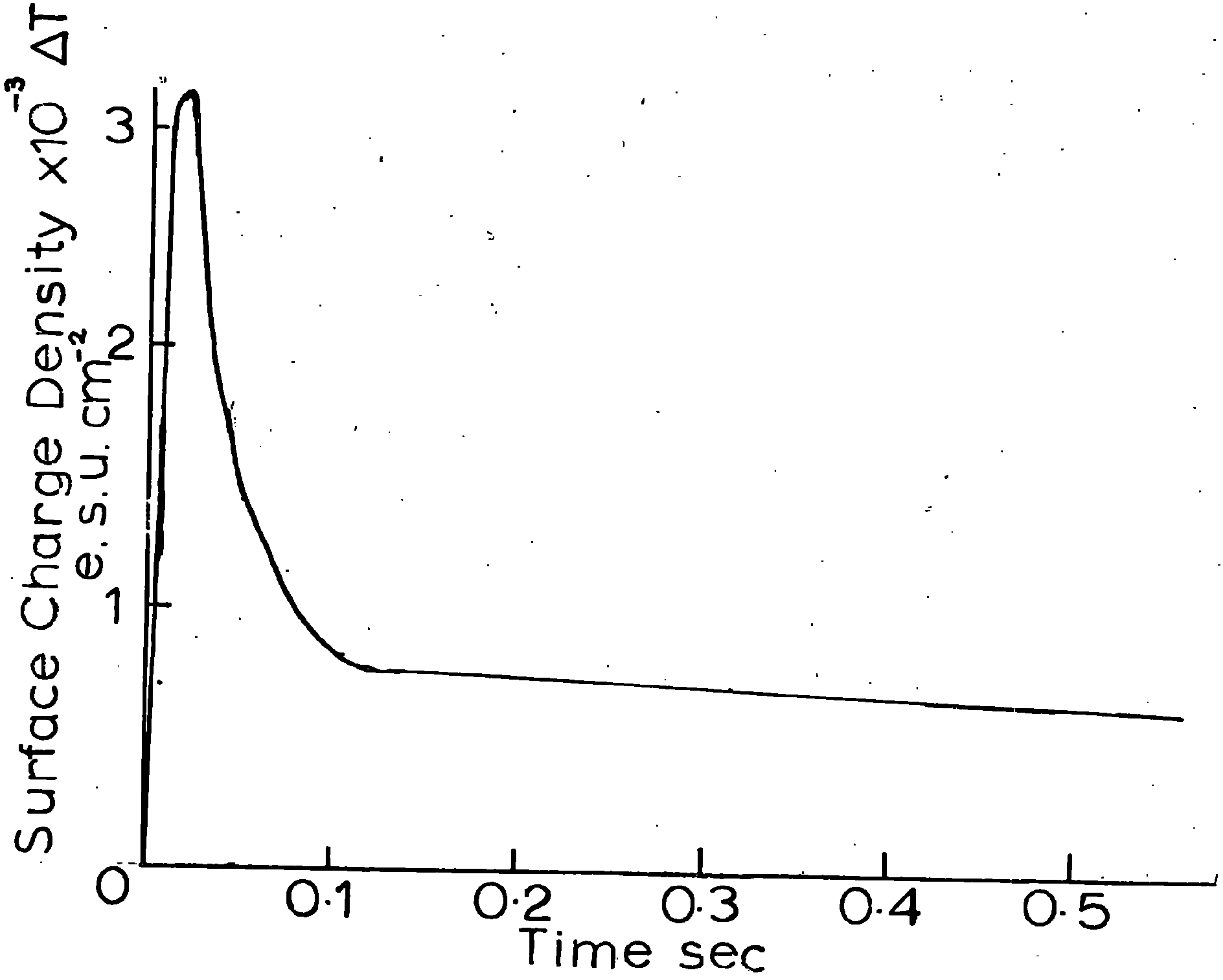


Fig. 5. The Charge Separated Across the Contact Plane of Two Pieces of Ice of Initially Different Temperature as a Function of their Time of Contact.

potentials near 0° C occurred mainly in the surface layers of the ice.

Jaccard (1963) took into account the effect of the Bjerrum defects on the thermoelectric power of ice, and allowed for the fact that the effective charge carried by each defect is less than the unit electronic charge. He showed that the predicted value would agree with that of Latham and Mason if the mobility ratio of the Bjerrum defects is 1.2.

Latham (1964), considering the charge separation due to non-uniform temperature gradients, suggests that the charge separated will be greater by a factor of two than the charge separated under conditions of uniform temperature gradient.

(b) Charge separation in water.

The principal ways in which a separation of charge may occur in liquids are spraying, splashing and bubbling. In contrast to charge separation phenomena in ice, these effects in water are due to the presence of impurities and the existence of an electrical double layer at the liquid-air interface. Loeb (1958) gives a detailed account of charge separation phenomena in liquids.

(c) Charge separation across a solid- liquid interface.

As stated in chapter 1, Workman and Reynolds discovered a charge separation effect across the ice - water interface of freezing dilute aqueous solutions. The potentials developed across the interface were found to depend on the type and concentration of the impurity in solution, and were only observed while freezing was progressing. Workman and Reynolds were able to show that the effect was ionic in nature by measuring the pH of the melted ice and the liquid solution after samples were partially frozen. The pH values of the frozen and non-frozen fractions were found to differ, thus showing that the ions of the impurity were being segregated. In general they found that the negative ions were incorporated into the ice structure although the NH_4^+ ion, which is isomorphous with the hydronium ion, was also incorporated into the structure. When ammonium fluoride was present the F^- ion was found to have a position of preference as it is strondly electro-negative and similar to the OH^- ion in structure. On the other hand the NO_3^- and CH_3COO^- ions were rejected by the ice on the freezing of solutions of the ammonium salts.

This phenomenon appears to be related to that of the rectifying action of semi-conductor materials, and the ice was in fact found to possess a slight rectifying action.

It also seems likely that the effect only occurs in the presence of impurities and not for pure water. Workman and Reynolds speculate as to a possible explanation by considering the water to pass through a transitional "colloidal" state on freezing as reported by Errera (1927). They suggest that it is reasonable to assume that the associated colloidal particles in the process of formation would avoid an increment in potential energy by incorporating ions of compatible form into their structure to form non-polar electrical domains rather than polarised ones. They suggest that the observed lack of any polarisation properties of ice might be due to collective action of the above type at the growing crystal face in such a way that the resonance pattern of the crystal units would be modified. They were able to obtain some evidence that the effect is related to a structural change on freezing by predicting similar effects for other compounds which undergo marked changes in bond structure on freezing. They investigated this in the case of phenyl salicylate and observed an effect which they believed to be similar to that occurring on the freezing of water.

(d) Charge separation across a solid-vapour interface.

When a specimen of ice evaporates in a current of dry air its outer surface will become colder than its interior. Applying the temperature gradient theory the outer surface should thus become positively charged. Evaporation of the outer surface would lead to a positive space charge in the airstream and a negative charge on the residual ice. Latham and Stow (1965) have investigated this effect and their observations will be discussed in chapter 6.

(e) Charge separation across a liquid-vapour interface.

Reshetov (1961) has suggested the existence of a mechanism by which water drops become charged by evaporation or condensation. He suggests that at the water-air interface a number of water molecules dissociate to form an ionic vapour of H^+ and OH^- ions. If the water drops are evaporating then the H^+ ions are preferentially reabsorbed and the drops acquire a positive charge. If they are growing by condensation then the OH^- ions are absorbed and the drops acquire a negative charge. There is as yet no direct experimental evidence to support this theory.

CHAPTER 3.

THE ELECTRIFICATION ON FREEZING OF SINGLE WATER DROPS.

1. INTRODUCTION.

It can be seen from chapter 1 that several of the proposed mechanisms of electrification in a thundercloud depend upon electrical effects associated with the freezing of water. This has led to several investigations into the electrification associated with the freezing of single water-drops. These investigations are described below.

2. THE PHYSICAL BEHAVIOUR OF WATER DROPS ON FREEZING.

The formation of spicules on the surface of freezing water was first investigated in the 1920's. In 1948 a mechanism for their formation was proposed by Dorsey. He suggested that as a volume of water freezes from the surface inwards, the pressure exerted on the liquid water becomes sufficiently high for it to cause a rupture in the ice surface. A jet of water is then ejected and freezes on the outside, the water continuing to be ejected along the tube formed until the pressure is equalised. The tube then freezes completely to form a spicule. Blanchard (1951) investigated the growth of spicules on freezing drops, of diameter 8 mm, which were freely supported in a vertical wind tunnel. By observing the rate of spicule growth he was able to verify the mechanism suggested by Dorsey.

Blanchard (1955) carried out further investigations into the freezing of water drops of diameter 8 mm, which were freely supported in a vertical wind tunnel. He found that the manner in which supercooled water drops froze depended on their temperature at the time of freezing. At temperatures above -4 or -5° C a shell of clear ice first formed on the bottom of the drop and then grew over the whole surface. The drop then froze inwards. On occasions, examination of a freezing drop showed the growth of thin planes of ice from the base of the drop, up through its interior, which accompanied the initial freezing. At temperatures below -5° C the freezing occurred nearly simultaneously over the entire surface of the drop, causing the drop to become opaque. Blanchard suggested that the opacity of the ice was due to the presence of small air bubbles which were formed as the air was forced out of solution as the water froze.

The production of ice crystals by the bursting of freezing water drops was observed by Mason (1956), and also by Langham and Mason (1958) who suggested that this might be an important mechanism in the ice-nucleus economy of supercooled clouds. In order to obtain further information Mason and Maybank (1960) carried out a detailed investigation into the splintering produced by the bursting of the drops on freezing. Drops having diameters in the range 30μ to 1 mm were suspended on fibres in a small refrigerated cell, the temperature and humidity of the cell being controlled. The drops were allowed to supercool to a particular temperature and then freezing was initiated by nucleation of the drop with either silver iodide crystals or small ice particles. The number of splinters ejected and the manner in which freezing progressed were carefully observed. They found that the manner in which the drops froze was dependent upon their degree of supercooling before nucleation. Drops which were supercooled to about -15° C were observed to freeze in two stages. The first stage was the rapid formation of an opaque outer shell of ice containing numerous tiny air bubbles, accompanied by a rapid rise in the temperature of the drop to 0° C. During the second stage the liquid interior of the drop gradually froze. During this stage liquid water from the drop interior seeped through the ice shell and froze on the surface. When drops were frozen at temperatures only a degree or so below 0° C a thin, transparent film of ice was observed to spread slowly over the surface of the drop. When the entire surface of the drop was frozen the freezing then progressed towards the interior of the drop, often with the formation of spicules. Observation of a number of drops showed that the frequency of spicule formation, shattering and splinter production decreases rapidly as the nucleation temperature is lowered, but is independent of drop size over the range investigated. Mason and Maybank suggested that this was due to the variation in the solubility of air in water with temperature. The solubility of air in water increases from 37 mg litre^{-1} at 0° C to 46 mg litre^{-1} at - 10° C. They suggested that the rapid increase in temperature of a freezing drop when nucleated at -15° C causes air to be forced out of solution and trapped in the ice as air bubbles. As further freezing of the drop progresses the pressure excess due to the freezing of the liquid interior is accommodated by compression of the air bubbles. At higher nucleation temperatures the ice shell forms more slowly and the dissolved air which is forced out of solution is lost to the atmosphere. The ice shell is thus transparent and mechanically strong. The excess internal pressure can thus only be relieved by the rupture of the ice shell, and, therefore, by the production of spicules and ice splinters. They were able to confirm this by observing the freezing of de-aerated drops. Drops nucleated at temperatures just below 0° C showed no difference in their behavious from that of aerated drops. However, de-aerated drops nucleated at -13° C showed greatly enhanced splinter production when compared with aerated drops. Mason and Maybank also found that the presence of impurities had no effect on the freezing behaviour of the drops except in the case of a 0.2 N solution of sodium chloride. Drops of this solution showed no distortion and produced no spicules and only a few ice crystals when nucleated at either -12° C or 0° C. This was put down to the fact that such a solution was observed to increase in overall volume by only one half of the amount associated with a similar volume of pure water when frozen.

3. THE ELECTRIFICATION OF WATER DROPS ON FREEZING.

(a) The work of Mason and Maybank (1960).

Following their investigation of the physical aspects of the freezing of single water drops, Mason and Maybank investigated the electrification caused by the freezing. The majority of their measurements were made on 1 mm diameter drops of doubly - distilled and de-ionised water. The drops were suspended from a fine, insulating fibre of polythene in the centre of a refrigerated cell, and between two parallel brass electrodes across which a steady potential difference was maintained. The drops were observed with a microscope while freezing, and any electrification of the drop was detected by observing the movement of the drops between the electrodes. The smallest detectable charge was 5×10^{-5} e.s.u.

Mason and Maybank found that no detectable charge was produced unless freezing was followed by shattering of the drops. They also found that shattering always produced a charge. They divided their results into two categories;

(1) those in which more than half of the original mass of the drop remained on the fibre after shattering; this they termed major residues.

(2) those in which less than half of the original mass of the drop remained on the fibre after shattering; this they termed minor residues.

Under the same conditions of freezing the major residues became negatively charged on average, while the minor residues became positively charged. The overall maximum charge observed was $- 7.2 \times 10^{-3}$ e.s.u., the smallest $- 0.11 \times 10^{-3}$ e.s.u. The overall average charge for 83 drops was $- 0.86 \times 10^{-3}$ e.s.u. The most frequent shattering and the greatest electrification was observed with 1 mm diameter drops nucleated near 0° C. Under similar conditions of freezing they found that drops of 0.35 mm produced charges smaller by a factor of two than drops of 1 mm diameter. However, insufficient data were obtained to establish any variation of electrification with size. They also found that drops of sodium chloride solution, frozen under conditions similar to those for pure water drops, ruptured less frequently and for those that did rupture the charging decreased with increasing concentration of salt; both shattering and electrification were totally inhibited when the concentration of sodium chloride exceeded 10^{-2} N.

Mason and Maybank suggested that the electrification was caused by charge separation along the temperature gradients produced in the ice shell during freezing according to the Latham- Mason theory. During the freezing of the drops a temperature gradient is set up across the ice shell as the outer surface attains the ambient temperature while the inner surface is maintained at 0° C as it freezes. Thus the outer surface of the ice will become positively charged, while the inner surface will become negatively charged. Any splintering of the outer surface of the ice shell under these conditions will carry away a positive charge, leaving the residue with a negative charge. If a major part of the drop is blown off the minor residue may be left with either a positive or a negative charge. On the basis of the temperature gradient theory Mason and Maybank calculated that the maximum charge separation for 1 mm diameter drops of pure water would be about 10^{-4} e.s.u. This is an order of magnitude less than some of their observed results. However, they point out that the effects of trace contaminants and frictional effects from the shattering of the ice are not known.

(b) The work of Kachurin and Bekryaev.

Kachurin and Bekryaev (1960) investigated the electrification on freezing of drops with diameters between 0.2 and 2 mm diameter over the temperature range -3 to -20° C. The drops were suspended on a fine wire eyelet in a temperature - controlled cell. The drops were at the focus of a microscope and the freezing of the drops recorded by filming through the microscope. The charge produced on the freezing drops was measured by connection of the supporting wire eyelet to either an electrometer or an oscilloscope. They found that significant electrification occurred only when shattering of the drops occurred on freezing. A reproduction of a typical oscillogram obtained by Kachurin and Bekryaev is shown in Fig. 6. From the film of the freezing they were able to show that the sharp positive increases (peaks 2, 3 and 4) were associated with the breaking off of negatively charged ice crystals, while the smooth negative increase in charging was associated with the ejection of streams of positively charged microscopic water drops. The first charge peak on the oscillogram was always positive, apparently being associated with the breaking off of a number of small ice crystals on the formation of fissures in the freezing ice shell.

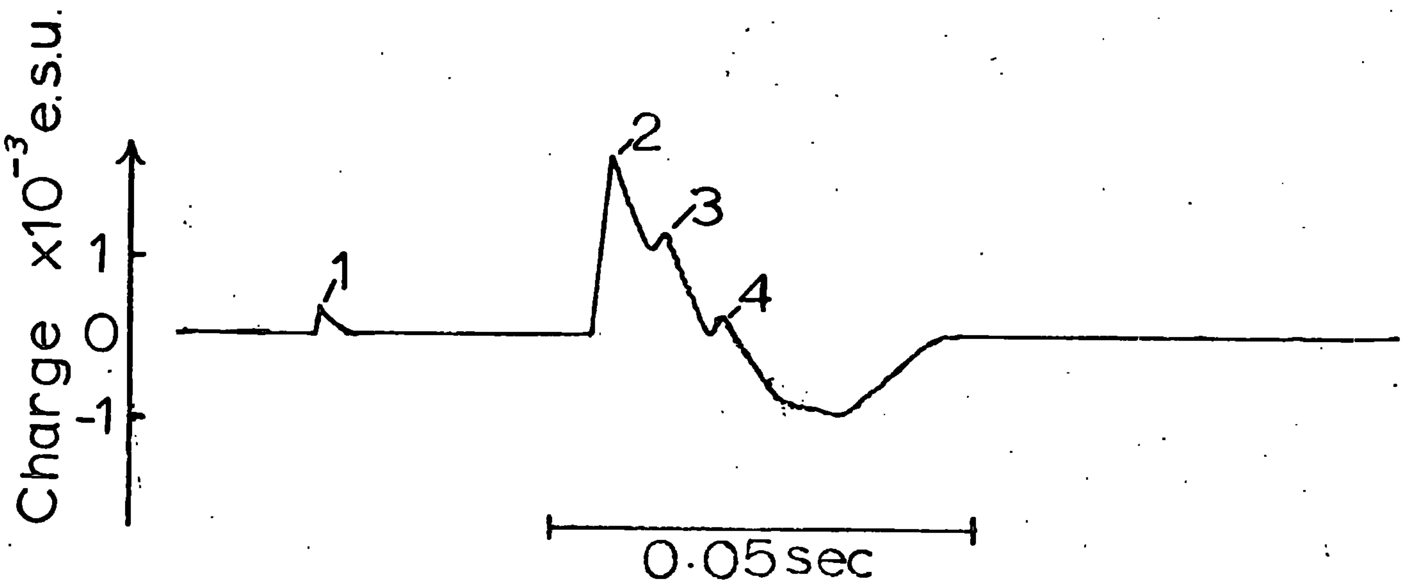


Fig. 6 Oscillogram of the Charge on a Freezing Water Drop.

Electrometer measurements of the net charges on the drop residues showed them to be less than the maximum charges on the fragments thrown off. The average value of the charge on 70 drops was $- 9.0 \times 10^{-13}$ C, or $- 2.7 \times 10^{-3}$ e.s.u. They did not observe any variation of the charge with varying drop size.

(c) The work of Evans and Hutchinson

Evans and Hutchinson (1963) investigated the electrification on freezing of water drops having diameters between 1 and 1.5 mm. The drops were suspended on an insulating fibre, made of "Durofix" adhesive, in a refrigerated cell in which a constant vertical temperature gradient was maintained. The drops were allowed to supercool to about $- 2^{\circ}$ C and then nucleated by a cloud of small ice crystals produced by the introduction of a small piece of solid carbon dioxide into the cell. After nucleation the drops were rapidly lowered to the $- 15^{\circ}$ C level of the cell. The freezing was then observed, and when complete the charge on the drop residue was measured by raising it into a Faraday cylinder which was connected to a vibrating reed electrometer. The charges detected ranged from $+ 11.3 \times 10^{-3}$ e.s.u. to $- 25.0 \times 10^{-3}$ e.s.u., the average charge value being $- 1.1 \times 10^{-3}$ e.s.u. The freezing of the drops was observed to be similar to that observed by Mason and

Maybank for drops nucleated at temperatures near 0° C.

Evans and Hutchinson divided their results into three classes according to the way in which fragmentation of the drops occurred.

(1) when the drop split in two, one portion being thrown off.

(2) when the spicule formed broke and was ejected almost immediately after formation.

(3) when the spicule was broken and ejected with the final freezing of the drop.

They suggested that in case (1) there would be a small quantity of liquid water at the centre of the drop when fragmentation occurred, in (2) the drop would be mainly liquid with liquid inside the spicule, and in (3) the spicule would contain little or no water. They considered whether the magnitude and sign of the observed charges was explicable in terms of the temperature gradient theory of Latham and Mason. Using the value of 5×10^{-5} dT/dx e.s.u. cm^{-2} calculated by Latham and Mason, they found that the maximum charge possible for an average drop would be Q max. $= 0.3 \times 10^{-3}$ e.s.u. However, of the 44 charges measured, 33 exceeded Q max, 13 by a factor of five or more.

Evans and Hutchinson consider that the signs of the charges observed by them were, on the whole, explicable in terms of the temperature gradient theory. The charges observed for drops which split were preponderantly negative. This would be expected on the basis of the temperature gradient theory, which assigns a positive charge to the outer surface of the ice, if an excess of the outer surface was thrown off. In cases where only the spicule is broken off, equal areas of the inner and outer surfaces would presumably be ejected, and thus the theory would predict an equal frequency of positive and negative charges. They reported 11 cases of positive charge and 18 of negative charge. In cases where the spicules were ejected with the final freezing of the ice it would be expected that the cold outer part would carry away positive charge, leaving a negative residue. In fact, 5 of the 6 residues of this type were positively charged. They also pointed out that the signs of the charges could also be explained in terms of the Workman - Reynolds effect. Referring to their classification of the types of freezing, methods (1) and (2) could involve the ejection of liquid water, leaving an excess of negative charge, while in method (3) the ejected particles would probably be mainly solid, leaving the positively charged liquid in the residue.

(d) The work of Stott and Hutchinson.

Following on the work of Evans and Hutchinson, further observations of the electrification of freezing drops were carried out by Stott and Hutchinson (1965). The apparatus used was similar in its essentials to that used by Evans and Hutchinson, and the experimental procedure was also similar, except that after shattering the drops were allowed to melt by raising them above the 0° C level of the refrigerated cell. The mass of the residue was then found by measurement of the size of the melted drop. The drops were formed from singly distilled water. The drop sizes were in the range 0.9 to 1.7 mm diameter, the average value being 1.3 mm. A total of 633 drops was observed, of which 118 shattered. In all but a few cases the drops were nucleated at - 1° C and then frozen at an ambient temperature level of - 15° C. The behaviour of the drops on freezing was found to vary from drop to drop. However, it was found that the types of freezing behaviour could be separated into quite well defined classes. After the first film of ice was seen to spread over the surface of the drop and begin to thicken, small irregularities developed on the surface of the drop. They suggest that this was due to water seeping through cracks in the ice shell and then freezing onto the surface. After this, one or more spicules or bulges were formed in all cases. The spicules were observed to be oval in cross-section, suggesting that they had been formed by the ejection and subsequent freezing of water from a crack in the ice shell as predicted by Dorsey. Stott and Hutchinson classified the spicules as being of two types. A "smooth spicule" always grew in a smooth, even way; after some seconds small bubbles formed inside it at the ice-water boundary, and moved into the drop or to the end of the spicule; finally a series of cracks spread through the drop and the water within froze quickly. Any shattering of the drop was observed to take place either just after the spicule was formed or while the cracks were spreading through the drop centre. "Bubbly drop spicules" were different in that as soon as the spicule began to grow, small air bubbles were released at the ice-water boundary in the main part of the drop. These spicules grew slowly with no cracking, and only a few broke. Presumably the air bubbles accommodated the pressure increase. On occasion a drop developed a bulge which did not grow into a spicule. As the growth of these bulges resembled that of bubbly drop spicules they were classified as "bubbly drop bulges". The drops were also classified according to the manner in which breaking occurred. A central break was one in which the drop broke through the centre. "Liquid" and "solid spicule breaks were those in which the spicule was broken off when it was still partly liquid, or all ice respectively. "Bubble" breaks were those in which spicules containing air bubbles were broken off. These classifications are shown in Fig. 7.

Of the 118 drops which were observed to shatter, 91 were found to have measurable charges. The charges ranged from - 17 x 10^{-3} e.s.u. to + 25 x 10^{-3} e.s.u. Stott and Hutchinson considered whether or not the observed charges were consistent with those to be expected on the basis of the temperature gradient theory. As pointed out by Evans and Hutchinson it would be expected that the different types of break as classified by them would have preferred signs of charge. Stott and Hutchinson found no evidence that the different types of residue had any preferred sign except in the case of bubbles, which produced only small charges. Stott and Hutchinson also calculated the maximum possible charge separation due to the temperature gradient effect. In order to allow for the enhancement of the charge due to part of the ice being warmer than - 7° C, and to the non-uniformity of the temperature gradient, they applied the correction factor of 6 proposed by Latham (1964). The predicted charge separation was thus equal to 0.3 x 10^{-3} dT/dx e.s.u. cm^{-2}. Taking their average drop diameter as 1.3 mm, and the average thickness of the ice shell as 0.3 mm at the time of breaking, they calculated the maximum charge separated to be 5 x 10^{-3} e.s.u. A similar calculation for the separation of charge for an average spicule

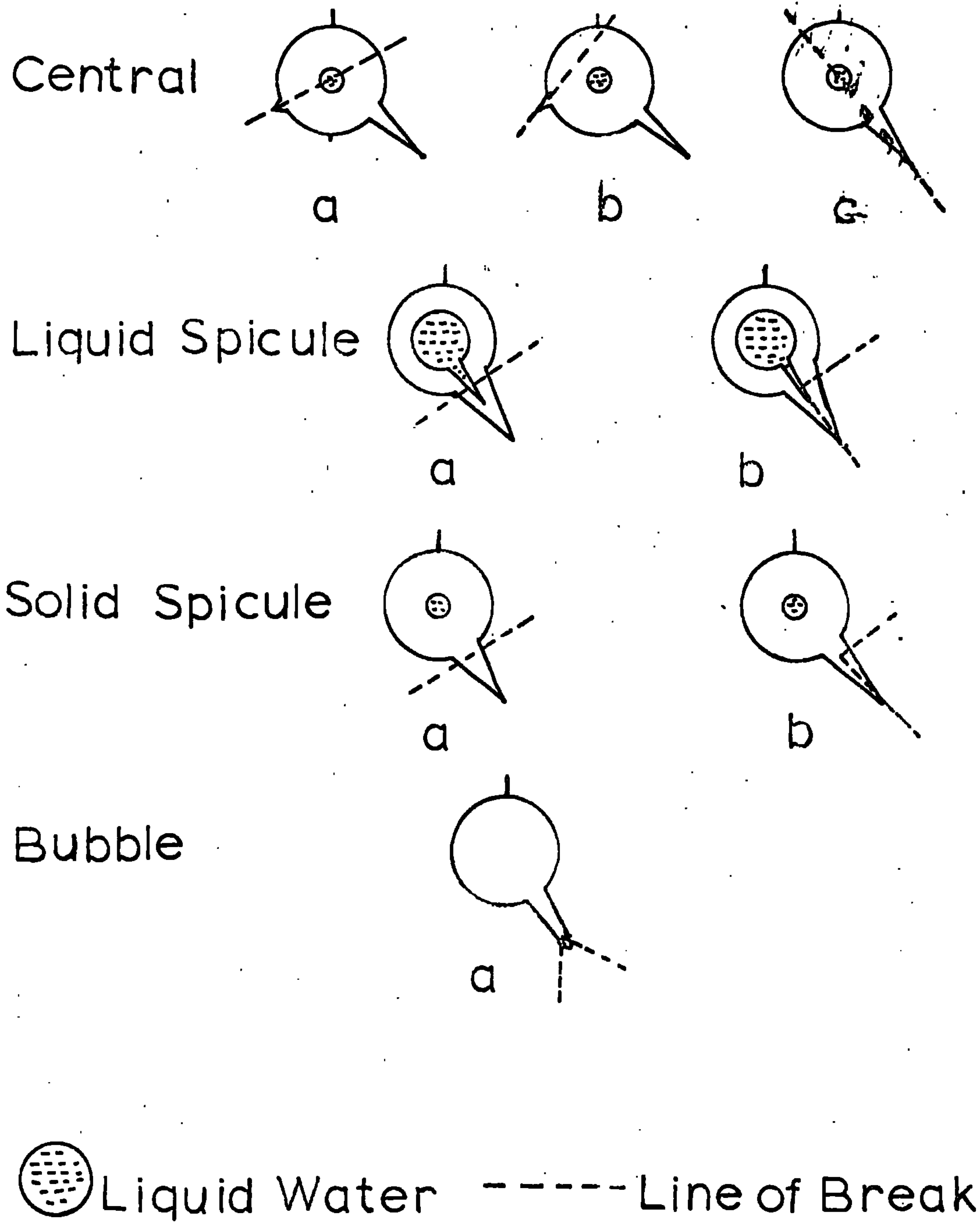


Fig. 7. Classification of the Types of Drop Break After Stott and Hutchinson.

of length 1.5 mm, average outside radius of cross-section 0.06 mm and thickness 0.03 mm, showed the maximum charge separated to be 6 x 10^{-3} e.s.u. Of the observed charges 13 were larger than this, and when allowance was made for the fact that only a proportion of the maximum charge would be removed on the ejected particles, it was seen that the theory as applied could not account for quite a number of the observed charges. They then investigated whether there was any tendency for positive residues to be produced by solid spicule breaks, and negative residues by liquid spicule breaks. They found the average charge produced on the residue to be $+(3.5 \pm 2.3) \times 10^{-3}$ e.s.u. for 19 cases of solid spicule breaks, and $-(1.6 \pm 1.0) \times 10^{-3}$ e.s.u. for 29 cases of liquid spicule breaks. When the results of Evans and Hutchinson were combined with those of Stott and Hutchinson the average values were $+(3.4 \pm 2.2) \times 10^{-3}$ e.s.u. (25 drops) and $-(1.4 \pm 0.9) \times 10^{-3}$ e.s.u. (49 drops) respectively. These differences are apparently significant. Stott and Hutchinson considered these results in view of the Kachurin and Bekryaev results, and suggested that a liquid spicule would tend to lose more water than ice on breaking, thus ejecting a net positive charge, while a solid spicule would eject an excess of negatively charged ice on breaking. They point out that this would be in agreement with the Workman - Reynolds theory as regards the signs of the charges.

4. APPLICATION OF FREEZING DROP RESULTS TO THUNDERCLOUD ELECTRIFICATION.

Mason and Maybank considered the quantity of charge which could be generated in a thunderstorm by the freezing of all the raindrops present. They considered the total rainfall to be 2.5 cm for a storm of area 10 km^2, the mean raindrop mass being 10 g and the average charge produced by freezing being 10^{-3} e.s.u., and calculated the maximum charge produced to be 8.3 coulombs. This is much less than the charge of 1000 C which is estimated to be present. They suggested, however, that the electrification accompanying the riming of soft hail might be caused by the splintering of cloud droplets on freezing.

The other investigators have not applied their observations to thundercloud electrification. However the value of the charges observed by them could only account for an increase in the charge production due to freezing raindrops by a factor of, at best, 5 times that calculated by Mason and Maybank.

There are doubts as to the validity of any attempt to apply the results of the laboratory investigations to the electrification of thunderclouds. In all cases it is uncertain whether of not the presence of the supporting fibre affected the behaviour of the drops. This is of particular importance in the work of Kachurin and Bekryaev where the heat conducted down the supporting wire would certainly have altered the local temperature gradients in the drop. Also, in all cases, the drops were frozen in stagnant air and it is possible that the freezing behaviour, and thus the electrification, may be considerably different for drops falling at their terminal velocity in air.

The results of Evans and Hutchinson and of Stott and Hutchinson may be questioned because of their use of solid carbon dioxide as a nucleating agent, which would give rise to excessive concentrations of the gas in their refrigerated cells. Dinger and Gunn (1946) found that the presence of excess carbon dioxide could neutralise the charging produced on the melting of ice. Also, Latham and Mason (1961) found that the presence of excess carbon dioxide could affect the charge separation due to a temperature gradient.

Further, their results would seem to be of only limited application in considering the thunderstorm electrification insofar as all of their observations were made on drops which were nucleated at - 1 or - 2° C, and it is not certain that the majority of raindrops or cloud drops are nucleated at such a high temperature. Certainly the majority of cloud droplets nucleated by accretion on a hailstone will be nucleated at lower temperatures than this.

5. THE PURPOSE OF THE PRESENT INVESTIGATION.

It was the purpose of the present investigation to observe the freezing of single drops which were freely supported, and to observe whether the freezing behaviour was different from that observed with suspended drops. It was also hoped to obtain further information concerning the effect of freezing behaviour on the electrification.

CHAPTER 4.

APPARATUS FOR THE INVESTIGATION OF SINGLE DROPS.

1. INTRODUCTION.

As stated in chapter 3 it was the purpose of this investigation to observe the electrification on freezing of drops which were supported without contact with anything solid.

2. CHOICE OF SUPPORTING SYSTEM.

Two ways of freely supporting water drops were considered:

(a) by electrostatic suspension similar to that used in Millikan's experiment.

(b) by means of a wind tunnel.

The electrostatic support system was rejected for two reasons:

(i) any variation in the position of the drop on freezing could be due to either a change of mass, due to shattering, or to a change in charge, and the effects would be indistinguishable.

(ii) in order to support a drop of 1 mm diameter carrying a charge of 10^{-3} e.s.u. the supporting electric field would have to be of the order of 150 Vcm^{-1}. Observations made in an electric field of this value would not necessarily be applicable to the freezing of drops in lower electric fields, and also, the effect of the initial charge on the drop would be difficult to determine.

For these reasons it was decided that supporting the drops by means of some kind of vertical wind tunnel would be most suitable. Two types of wind tunnel were considered:

(a) one with uniform cross-sectional area as used by Blanchard (1951), for example.

(b) one with specially contoured cross-sectional area as used by Kinzer and Gunn (1951).

Blanchard observed that when water drops supported in a wind tunnel froze rapidly they became unstable and oscillated out of the supporting air flow horizontally. As symmetrical objects supported in a wind tunnel of the type used by Kinzer and Gunn are constrained to remain on the axis of the airflow it was decided to use this type.

3. THE SUPPORT TUBE.

Initial tests were carried out using a tapered glass tube which was drawn by hand. This tube varied in internal diameter from 2 to 6 mm over a length of approximately 4 cm. Drops of diameter about 4 mm were inserted into the tube by means of a hypodermic syringe and supported on the airflow through the tube. When the air flowing was cooled the drops were observed to begin freezing. However, when approximately a third of the drop being supported was frozen it became unstable and oscillated from side to side till it struck the tube wall. It was thought that this loss of stability might be due in some measure to irregularities in the taper of the tube. Because of this it was decided to construct tubes of a more uniform taper. In order to do this a hexagonal carbon mandril was made and several glass tubes drawn with its use. These tubes were tapered from an internal diameter of 3 mm up to 6 mm over a length of 9 cm. At the narrow end of the bore the tube was flared out to form a 5 cm length of 5 mm bore straight tubing for connection of the air supply. One of these tubes is shown in Fig.8. Observations of the freezing of drops using these tubes showed that the drops were mechanically more stable in these tubes. However, the taper of the tubes was still not completely uniform owing to irregularities in the carbon mandril. For this reason, and also to aid in the measurement of charge as discussed later, tubes were machined out of brass with the angle of taper closely controlled. It was first necessary to make a special tapered reaming tool. This tool was made by machining a steel rod until it had the required angle of taper and dimensions. Slightly less than half of the tapered section of the rod was then ground away so as to leave it with a D-shaped cross-section. This was hardened and used to ream out the tubes. The internal dimensions of these tubes were the same as those of the glass

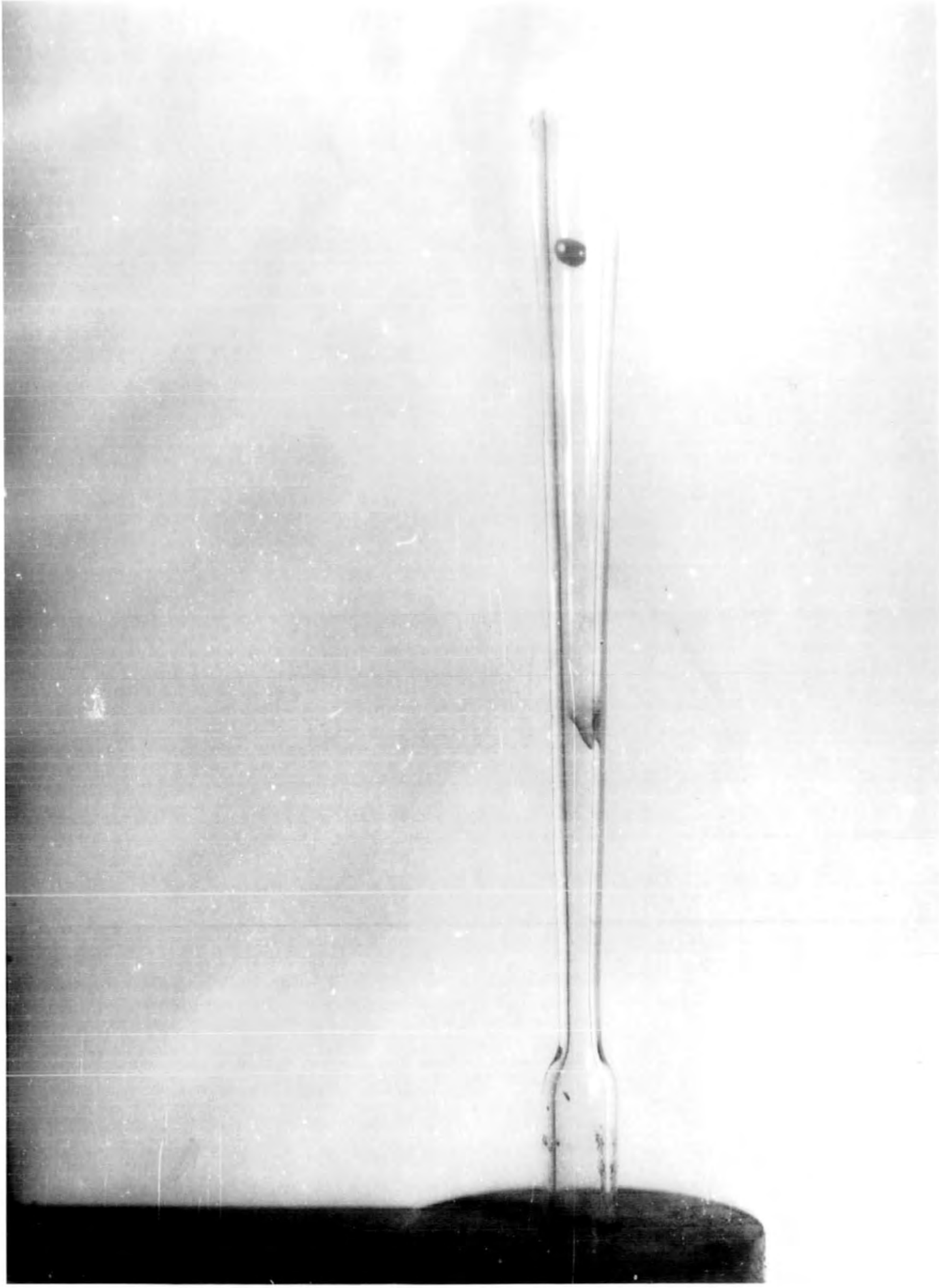


Fig. 3. A Coloured Water Drop Supported in a Glass Support Tube.

tubes, the lower end being machined so as to allow for the connection of the air supply. This is illustrated in Fig. 9. These tubes were found to be satisfactory in use. However, the freezing drops were still observed to become unstable when approximately one third of their volume had frozen. This was thought to be due to the loss of fluidity of the drop, and will be discussed further in Chapter 5.

4. THE AIR SUPPLY SYSTEM.

Initially the air supply was provided by a blower fan having a 3" diameter impeller and a ¾" diameter outlet nozzle. However, as discussed later, it was found necessary to use a small bore cooling system and this exerted too great a resistance to the air flow for the fan to overcome. Because of this cylinders of compressed air were used. However, difficulties arose in their use. The cylinders were supplied containing air at a pressure of 2500 p s i and the flow was regulated by means of a reducing valve, British Oxygen type S120.OG, by which the output pressure could be varied over the range 0 to 100 p s i. In use the output pressure was set at approximately 5 p s i. Under these conditions the air flow was found to be subject to random fluctuations of up to 15%. It was thought probable that these fluctuations were due to the inadequacies of the regulating valve when operating under these conditions. In

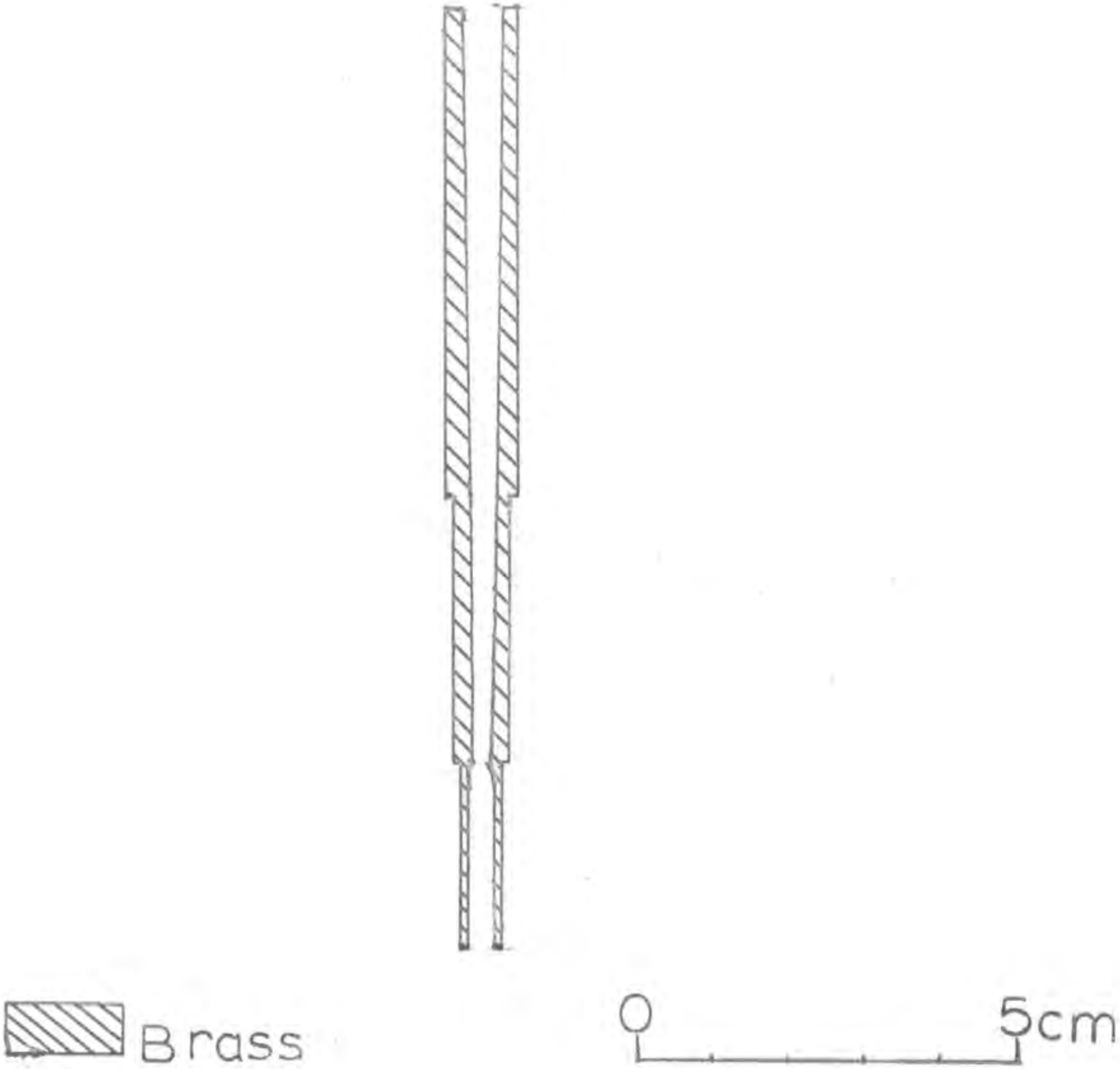


Fig. 9. Diagram of a Brass Support Tube.

order to overcome the fluctuations a length of 1mm bore glass tube approximately 5 cm long was inserted in the air line immediately following the regulating valve to act as a choke. It was then necessary to increase the output pressure to approximately 40 p s i in order to obtain the required flow rate. The flow rate was found to be constant and continuously variable over the range 0 to 15ℓ min^{-1}.

Measurements of the space charge present in the air supply were carried out using the space charge collector described by Groom (1966). Under the conditions where the air flow was fluctuating what appeared to be pulses of space charge were observed which corresponded to the fluctuation in the flow rate. When the air flow was stable this was not observed. It was found that these apparent space charge pulses were probably due to piezolectric effects in the space charge collector, caused by the pressure fluctuations of the airflow.

It was also found that quantities of water condensed out of the air following its adiabatic expansion on leaving the air cylinder. In order to avoid the transmission of this water through the cooling system it was frozen out in a water trap.

After the above precautions had been taken the air cylinders were found to be a satisfactory source of the air supply. Later in the course of the experiment a small air compressor, Broom and Wade Handyair 3, became available and this was then used as the air supply. The glass capillary was left in the airline to remove fluctuations due to the compressor action, and the space charge collector was also kept in the airline to filter out oil and dust particles from the compressor. The flow rate was again regulated by the reducing valve, and was measured by a Rotameter flowmeter which covered the range 0 to 10 ℓ min^{-1}.

5. THE DROP PRODUCER.

It was intended to investigate the behaviour of drops of diameter 1 mm so as to allow comparison of the results with those of previous workers. A drop producer was accordingly constructed similar to that of Levvy (1947). In this apparatus drops of the required size are formed on the tip of a hypodermic needle and then detached by an airflow which is co-axial with the needle. The drop producer worked satisfactorily over the range of drop sizes 0.5 to 3 mm diameter. However, it was not found possible to balance the air flow through the support tube with that around the needle so as to float the drops on the supporting air stream. Kinzer and Gunn (1951) produced drops by means of a fine glass capillary which was sealed into the top of their support tube. They made observations on the drops by allowing them to fall out of the bottom of the tube into a collector. In the present investigation the lower end of the support tube was connected to the air supply and so the top of the tube had to be left open to allow for removal of the drops. Because of this the drops were introduced into the air stream by hand, using a hypodermic syringe with a No.20 gauge needle. This limited the size of the drops to between 3 and 5 mm diameter.

6. THE TEMPERATURE CONTROL APPARATUS.

In view of the proposed use of the blower fan as the source of the air supply in the early stages of the investigation several attempts were made to design heat exchangers which would satisfactorily cool the air stream to -20° C while presenting little resistance to the air. These attempts were not succesful and so will not be discussed further. It was decided that it would be necessary to use a heat exchanger with a high resistance to the air and, as already stated, it was for this reason that cylinders of compressed air were used as the source of the air supply. Helices were constructed of 8 mm bore glass having 10 turns in a length of 10", the radius of the coils being 1". Two of these helices were connected in series and submersed in methylated spirits maintained at a temperature between -35° C and -40° C in a refrigerated tank. Using this arrangement of heat exchangers it was found that the air could be cooled to -23° C at a flow rate of 10 ℓ min^{-1}. As stated earlier considerable quantities of water were present in the air supply from the cylinders and this water was found to freeze in the heat exchangers and block them. To overcome this problem the air was passed through a water vapour trap before passing through the heat exchangers. Liquid nitrogen was used to cool the vapour trap but it was found that oxygen and carbon dioxide were frozen out of the air as well as the water. Because of this the water trap was cooled in the tank of methylated spirits. Small quantities of water were found to pass through the trap and then freeze in the heat exchangers. However, these quantities were found to block the heat exchangers only after several hours, and the heat exchangers were cleared of all ice at the beginning of each day's observations to prevent blockages developing. As the degree of cooling produced by this arrangement was not variable the air was passed over a small electric heating coil after passing through the heat exchangers. The air temperature was adjusted by varying the supply of current to the coil by means of an Ironstone variable transformer, the temperature being measured by copper-constantan thermocouples.

7. CHARGE MEASUREMENT.

As all the drops whose freezing was observed during testing of the support tube had become unstable and collided with the wall of the support tube it was decided to use the support tube as the charge collector. This is one reason for preferring the use of the electrically conducting material brass. This arrangement had the advantage that any change in the charge of the drop could be observed continuously. The arrangement of the support tube is shown in Fig. 10. The support tube was supported vertically in polytetrafluorethylene mounts which insulated it from earth. The tube was closely surrounded by an earthed brass cylinder and enclosed in an earthed aluminium box. The tube was connected to the input of an Ekco vibrating reed electrometer, type N616B. The electrometer output was connected to a fast-writing pen recorder, the Watanabe Mini-writer 201-L. In use the electrometer input was short-circuited during insertion of drops into the tube, and then the $10^{10}\Omega$ input resistor was selected. The observed noise-level was seen to fluctuate up to 0.5 mV while using the $10^{10}\Omega$ input resistor. The quantity of charge was found by measuring the pulse produced on the recorder output and instrument noise-level set the minimum detectable charge as 0.15×10^{-3} e.s.u.

8. THE LAYOUT OF THE APPARATUS.

The layout of the apparatus is shown in Fig.11. The support tube was mounted inside the refrigerated cell on an earthed aluminium plate above the methylated spirits. The lead from the support tube to the electrometer head

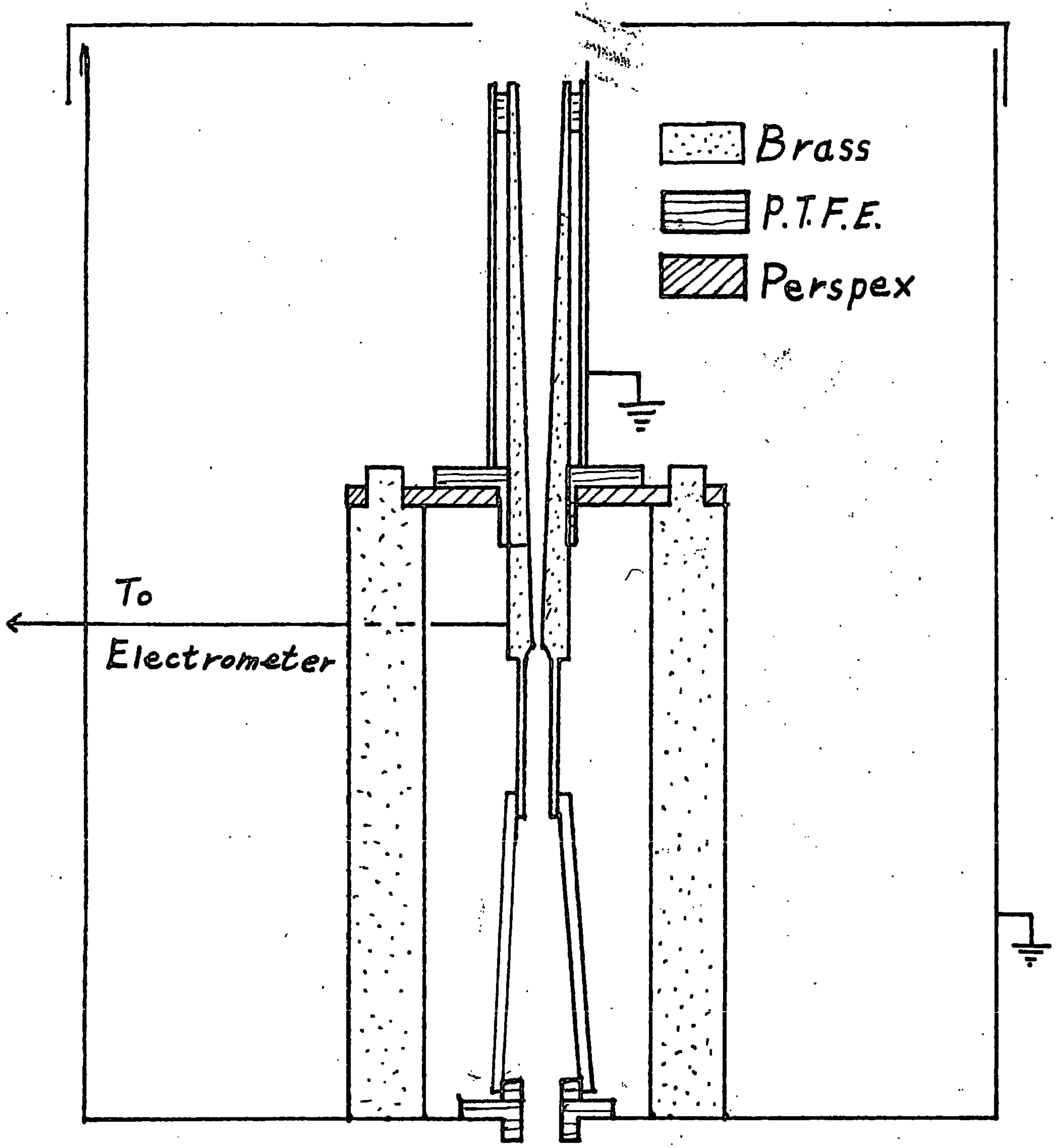


Fig. 10. The Support Tube and Charge Collector

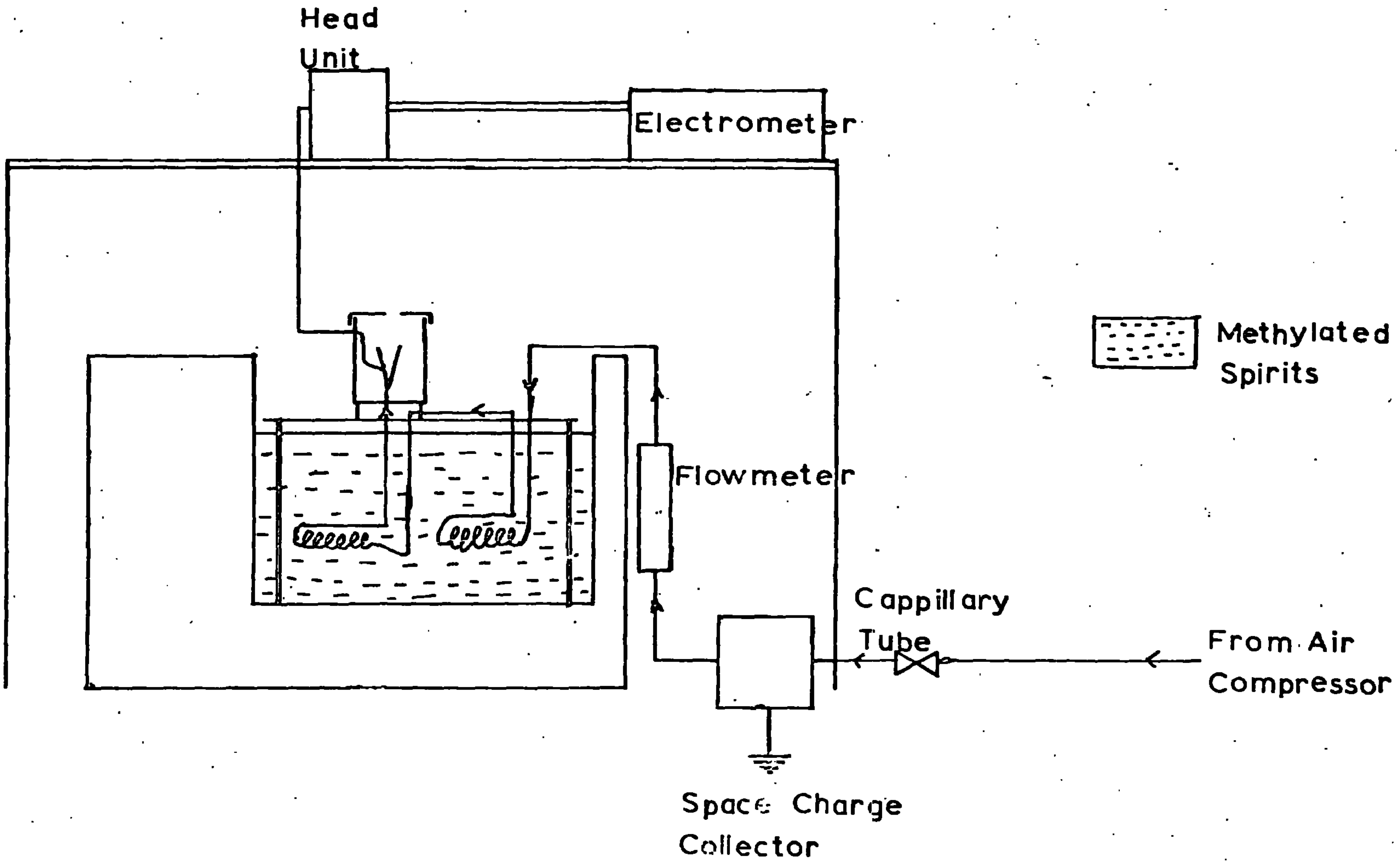


Fig. 11. Diagrammatic Experimental Arrangement

unit was rigidly fastened along its length so as to minimise the effect of piezoelectric effects due to vibration. For the same reason the head unit was mounted on a rigid freestanding rack. A low-power telescope was mounted above the support tube for observation of the drops. A tape-recorder was also mounted on the rack so that observations on the drops could be recorded. The compressor unit was sited outside the laboratory so as not to take in any excess carbon dioxide due to the occasional use of cardice in the laboratory.

CHAPTER 5.

EXPERIMENTAL PROCEDURE AND RESULTS.

1. EXPERIMENTAL PROCEDURE.

Basically the experimental procedure was to float a drop in the support tube at a particular temperature and then to observe the time lapse before the commencement of freezing, the physical behaviour of the drop on freezing and its electrification.

Before the drop was introduced into the support tube the flow rate of the air supply was set at the desired value as found previously by experiment. The temperature of the air was then adjusted by means of the heating coil, no observations being taken until the temperature had stabilised. When the air flow conditions were steady the thermocouple junction was removed from the mouth of the support tube and a drop introduced by hand, using the hypodermic syringe. Once the drop was floating freely in the airflow a stopwatch was started and the electrometer input setting changed from the "short circuit" to the $10^{10}\,\Omega$ setting. The size and freezing behaviour of the drop were observed through the low-power telescope and the observations spoken into the tape-recorder microphone. At the end of each observation the air flow conditions were checked. The drop residue was then removed from the wind tunnel ready for the next observation. Successive observations were identified on the tape-recorder and the pen-recorder by numbers. The following information was obtained for each drop:-

air temperature; drop size; the time for which the drop was supported before freezing commenced; the physical behaviour of the drop on freezing; and its electrification.

2. CALCULATIONS.

The temperatures quoted in Appendix 1 are the dry-bulb temperatures of the airflow, and before considering the results of the investigation it is necessary to find the temperature of the drops on freezing. Kinzer and Gunn (1951) have shown that an evaporating water drop will in time attain the wet-bulb temperature of the surrounding air-stream. To find the temperature of the drops on freezing it is necessary to know their thermal relaxation-times and the wet-bulb temperature of the air. The initial temperature of the drops on insertion into the support tube is uncertain as they were cooled during their growth on the hypodermic needle. Experience showed that they were probably at around 0° C as they tended to freeze on the needle if their growth was slower than usual. Because of the uncertainty in the temperature only approximate calculations have been made of the drop temperatures on freezing.

CALCULATION OF THE DROP TEMPERATURES ON FREEZING.

(i) The wet-bulb temperature of the air.

The lowest temperature attained by the air was -22° C and at this temperature the air was fully saturated. Extrapolation of the values of the weight of water present in unit volume of air given by Kaye and Laby (1958) show that approximately 1 gm m^{-3} of water is present. As the volume of the air changes by less than 1% over the temperature range investigated this may be taken as the amount of water present over the temperature range. The relative humidity of the air at various temperatures is then as shown in Table 2.

TABLE 2. Relative humidity of the air at various dry-bulb temperatures.

Dry bulb temperature	Relative humidity
0° C	20%
-5° C	26%
-10° C	44%
-15° C	50%
-20° C	100%

The wet-bulb temperature of the air is found by extrapolation from the values of the variation of wet-bulb depression with relative humidity given by Kaye and Laby. The dry and wet-bulb temperatures of the airflow are shown in Table 3.

TABLE 3. Dry and wet-bulb air temperatures.

Dry-bulb temperature	Wet-bulb temperature.
0° C	-5° C
-5° C	-8° C
-10° C	-12° C
-15° C	-16° C
-20° C	-20° C

(ii) The thermal relaxation-times of the drops.

Kinzer and Gunn (1951) have shown that the thermal relaxation-time of a freely falling water drop varies with size in approximately the manner shown in Fig.12, and that it decreases with decreasing humidity. Reference to Fig. 12 shows that the relaxation-time varies from about 4 to 7 seconds over the drop range of drop diameters from 3 to 5 mm. Approximate values are shown in Table 4.

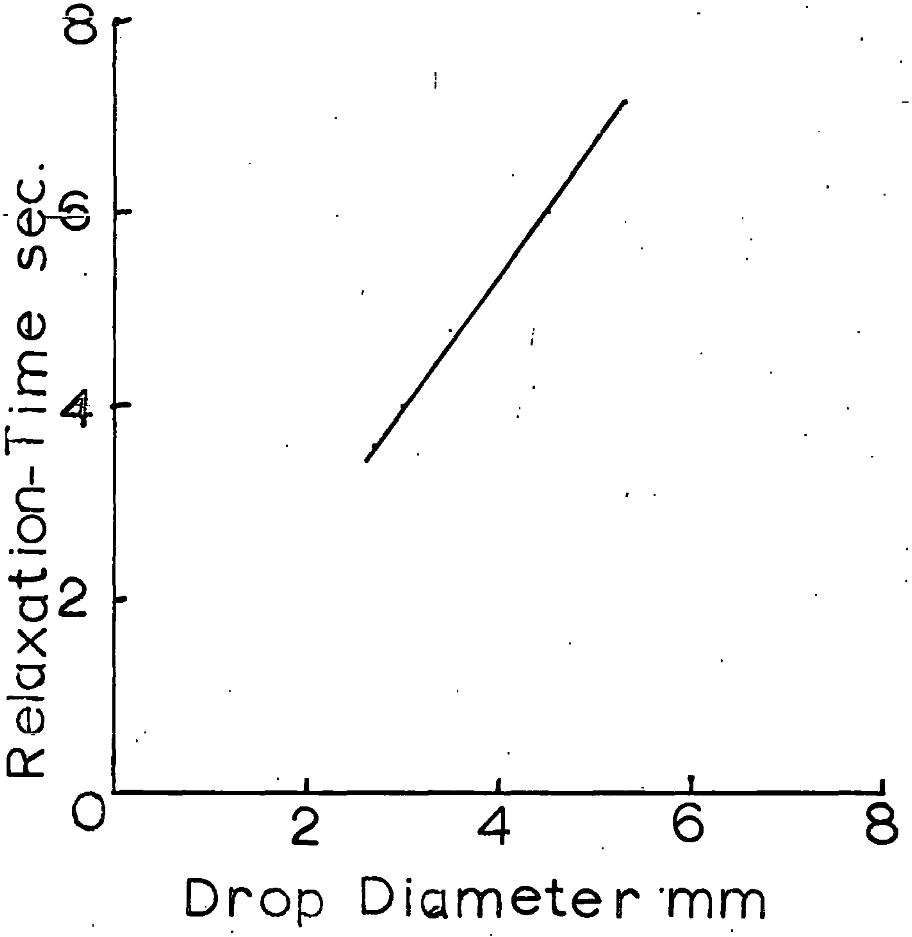


Fig.12. Variation of the Thermal Relaxation-Time of Evaporating Waterdrops with Size.

TABLE 4. Thermal relaxation-times of drops.

Drop diameter	Relaxation-time
3 mm	4 sec.
3.5 mm	4.8 sec.
4.0 mm	5 sec.
4.5 mm	6 sec.
5.0 mm	7 sec.

The calculated temperatures of the drops on freezing are shown in Appendix II.

3. RESULTS OF THE INVESTIGATION.

Observations were made on approximately 40 drops during testing of the apparatus, and detailed observations were made on 150 drops. The detailed observations are given in Appendix 1 and the calculated temperatures of the drops on freezing in Appendix II. Of these 150 drops Nos. 1-50 were of tap water, Nos.51-100 of de-ionised water, and Nos.101-150 of melted snow. The observations on the physical behaviour of the drops and their electrification will be considered separately.

(a) The physical behaviour of the drops.

During the testing of the apparatus observations were made on drops freezing at temperatures just below 0° C. The manner in which the drops froze was seen to depend on whether or not they were rotating in the airflow about a horizontal axis. When the drops were rotating fine ice crystals were seen to grow into the drop from the surface and to be uniformly spaced over the surface. When approximately half of the bulk of the drop had frozen it became unstable in the airstream and oscillated into the wall of the wind tunnel. If the drops were not rotating then ice crystals were observed to grow up through the drop from the base. This was followed by progressive freezing of the base from the drop upwards. Again, when half-frozen the drop became unstable and collided with the wall of the tube.

The observations given in Appendix 1 were all carried out on drops which were floated in the brass support tube. The observations cover drops in the diameter range 3 mm to 5 mm diameter, and the range of air temperature from -5° C to 22° C. No variation in the behaviour of the drops was observed over this range of sizes, or between the different types of drops. It was found that the manner in which the drops froze could be classified according to the temperature of the drops when freezing commenced.

These two classes are:

(1) drops freezing at temperatures above -10° C:

(2) drops freezing at temperatures below -10° C.

When the drops froze at temperatures above -10° C the freezing was observed to progress uniformly from the base of the drop upwards. When one-third of the drop was frozen it became unstable in the airstream and oscillated until it collided with the wall of the wind tunnel. The liquid portion of the drop then spread over the tunnel wall and froze onto it. When the drops froze at temperatures below -10° C a thin shell of ice froze rapidly over the surface of the drop except for a small, approximately circular area at the top of the drop. This area was typically 1 mm in diameter. A few tenths of a second after the formation of this shell the drop became unstable and moved horizontally until it hit the wall of the wind tunnel and adhered to it. The freezing of the drop then progressed uniformly from the base upwards. When approximately half of the drop had frozen air bubbles were seen to be forced out of solution. As the freezing continued chains of air bubbles were seen to be trapped in the ice, the bubbles being of the order of 0.2 mm in diameter. These chains of bubbles followed the curvature of the surface of the drop. In the final stages of freezing water was extruded through the hole in the ice shell. This water froze to form a bulge, giving the frozen drop the appearance of an onion. Close examination of this bulge showed it to contain trapped air bubbles, although where the base of the bulge joined the main part of the drop the ice was generally transparent. On occasions a very thin layer of ice formed over the hole in the shell before the extrusion of any water. This layer of ice was almost immediately broken by the extrusion of water, with no visible ejection of ice splinters or water drops. Freezing then continued as before.

It was noted that none of the drops which were observed while floating in the brass support tube appeared to rotate at all.

(b) The electrification of the drops.

None of the 150 drops whose electrification was observed were found to have any charge within the limits of measurement of the apparatus.

4. DISCUSSION OF THE RESULTS.

It was stated in Chapter 4 that all of the drops which were observed during freezing became unstable in the airstream before freezing was complete and hit the wall of the support tube. Blanchard (1955) quotes McDonald as pointing out that the centre of gravity of a falling water drop is above the upward acting centre of pressure for the aerodynamic forces on the drop. The drop remains in equilibrium by adjusting its shape continuously so as to prevent the above couple from acting. However, when the drop can no longer change its shape, due to the formation of ice, this couple begins to act and causes the drop to oscillate in the airstream. It seems likely that this is the reason for the drop instability in the present investigation. This would seem to be borne out by the observation that some of the drops were seen to change shape in an oscillatory manner when first introduced into the supporting airstream. These oscillations died out after a few seconds and the drops became stable. It seems that these oscillations were due to the drop changing shape so as to prevent the above couple from acting.

The physical behaviour of the drops on freezing is somewhat different from that observed by previous workers. As stated in chapter 3 previous investigations have shown that the freezing of drops at temperatures near to 0° C commenced with the slow growth of a shell of clear ice over the surface of the drop. This shell then thickened. In the present investigation the formation of this shell of ice was not observed, the freezing being seen to progress from the base of the drop and uniformly throughout it.

Blanchard (1955) found that for drops at temperatures below -5° C the freezing commenced by the very rapid formation of an opaque ice shell over the entire surface of the drop. Freezing then progressed by the thickening of the ice shell. Mason and Maybank(1960) observed a similar pattern of freezing behaviour for drops at temperatures of about

-15°C. In the present investigation the ice shell did not cover the entire surface of the drop, and after its formation freezing progressed uniformly from the base of the drop.

It seems probable that the differences in behaviour of the drops in this investigation as compared with the behaviour of drops on fibres is due to the different heat transfer conditions. Suspended drops will lose heat mainly by conduction to the surrounding air while drops which are supported by an airflow will lose heat by conduction and also by evaporation. Further, the heat loss from a suspended drop will be uniform over its surface while that from a freely supported drop will depend on the pattern of the surrounding airflow. The growth of ice from the base of the drop upwards is explicable if the rate of transfer of heat is greatest at the base of the drop. Similarly, the existence of the unfrozen portion of the drop's surface is explicable if the rate of heat transfer is least at this point. Spilhaus(1947) has determined the pressure distribution around a freely falling sphere and found it to be as shown in Fig.13. It can be seen from Fig.12 that the flow streamlines around the drop are concentrated at the base and sides while the top of the drop is in a region of stagnant air. Applying this picture to a water drop it is seen that the rate of heat transfer, which depends on the local air motion, is greatest at the base of the drop and least at the top. It should also be noted that convection currents will exist in the drop and these will also

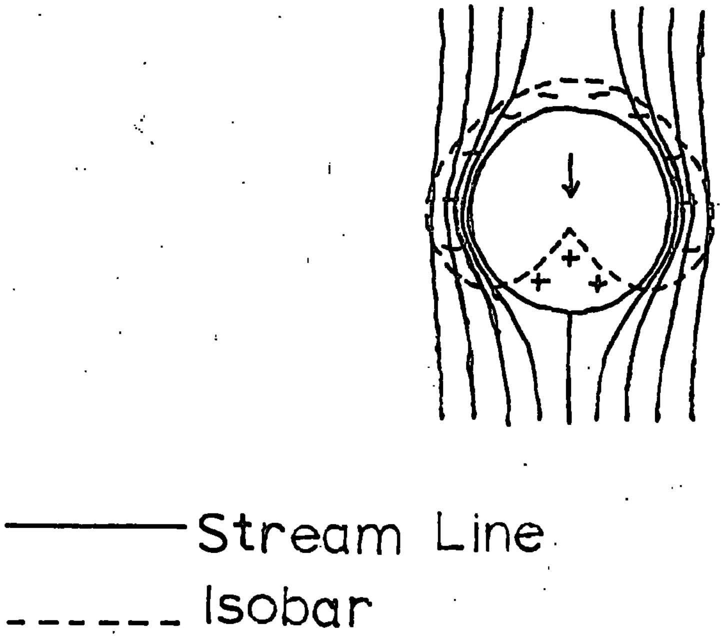


Fig.13. Pressure Distribution around a Sphere.

keep the top surface warmer than the remainder.

It is difficult to see why the behaviour of the drops was different in this investigation from that of Blanchard. The most probable explanation would seem to lie in the different airflows around the drops, and thus in the different conditions of heat transfer.

It is not known why the freezing behaviour of the drops changes in such a marked way with temperature. It is unlikely that the nucleating agent exerts any effect as the nuclei in this experiment were ice crystal fragments blown off the deposit on the walls of the heat exchanger and thus presumably always of a similar nature.

The results of the investigation into the electrification of the drops are significantly different from those of previous workers, as no electrification was found within the limits of detection of $\pm$ 0.15 x 10^{-3} e.s.u. Frequent tests were made to ensure that the charge measuring apparatus was functioning correctly and these are described in Appendix III. As previous workers have shown that electrification occurs only when the drop shatters or splinters on freezing this is understandable as no breaking of the drops was observed. Before considering the results further it is necessary to see if they could have arisen by chance. Previous workers have found that the overall probability of a drop breaking on freezing is about 0.2. Assuming this to be the probability a χ^2 test may be carried out on the results. From this test it is found that to better than the 0.1% confidence level the results could not have arisen by chance. Thus the difference of these results from previous ones may be taken to be significant.

In applying these results to the electrification of thunderclouds it must be remembered that after the initial stages of freezing the drops were no longer freely supported and it is possible that the final stages of freezing might be significantly different if they occurred while the drops were still freely falling. This would particularly be expected in the case of drops freezing at below -10° C when rotation of the freezing drop might lead to the entire surface freezing. This would lead to splintering of the drop as the freezing progressed. It should also be remembered that these results hold for drops much larger than the cloud drops present in a thundercloud. However, if these results are taken to be applicable to cloud drops in a thundercloud two conclusions may be drawn.

First, it seems unlikely that drops freezing at above -10° C would contribute to the ice-nucleus economy of the cloud, unless rotation of the drops caused the formation of a complete ice shell before the centre of the drops froze.

Secondly, it may be speculated that the electrification of riming hailstones could be temperature dependent. Drops which impinged on the hailstone at temperatures above -10° C would freeze slowly and the liquid water might spread over the surface of the hailstone. Further freezing would lead to the formation of glaze ice. Drops which impinged at temperatures below -10° C would maintain their spherical shape because of the rapid formation of the covering ice shell. If the entire surface of the drop was frozen over subsequent freezing would lead to breaking and electrification. This would give rise to an opaque rimed surface. It is interesting to note that this might be the cause of the negative charge centre of the thundercloud being at the -10° C if the different types of freezing produced electrification of different signs. This will be discussed again later.

CHAPTER 6.

THE ELECTRIFICATION OF HAIL.

1. INTRODUCTION.

Because of the importance of the role of solid precipitation in many of the theories of thunderstorm electrification, many workers have investigated the electrification of hail by various methods. This chapter reviews these investigations.

2. THE ELECTRIFICATION PRODUCED ON MELTING.

The electrification produced on melting is of importance for ice crystals and snowflakes as well as for hailstones.

Dinger and Gunn (1949) found that when samples of pure ice were melted in an airstream the water acquired a positive charge of 1.25 e.s.u. gm^{-1} of water melted, while an equal and opposite charge was carried away in the airstream. The charge was found to be sensitive to the presence of impurities, small quantities of which neutralised the charging. They also found that the presence of dissolved gases was essential for the charging to occur and suggested that the charging was connected with the cataphoresis of the gas bubbles when they were released from solution.

Mathews and Mason (1963) repeated Dinger and Gunn's experiment and also observed the electrification on melting using two other experimental arrangements. In no case did they observe any charge. Dinger (1964) suggested that this was because the charge effect is sensitive to the presence of carbon dioxide, and large quantities of the gas were present during the Mathews and Mason experiments.

Kikuchi (1965) performed experiments on the melting of natural snow crystals and on samples of ice containing various concentrations of air bubbles. The snow crystals were found to become positively charged on melting. The water was also found to become positively charged when the ice samples melted, and it was found that the quantity of charge acquired by the water was proportional to the concentration of air bubbles over a wide range. Kikuchi concluded that the air bubbles were responsible for the charging.

Magono and Kikuchi (1963, 1965) studied the melting of both natural and artificial snow crystals and found that they acquired a positive charge on melting. The charge increased with the size and complexity of the snowflake and they concluded that the charge was due to the release of air bubbles trapped in the ice. On average, a snow crystal melting to form a drop of diameter 150μ acquired an average charge of 2 x 10^{-4} e.s.u. If this result is generally applicable to snow, then the process might be a powerful generator of charge in snowstorms.

MacCready and Proudfit (1965A) made measurements on the charges of hydrometeors in and below thunderclouds. They found that the charges on graupel and hail within the supercooled regions of the cloud were generally large and positive. Below the cloud the positive charge was found to decrease steadily until the hydrometeors were at the 0°C level. The charge then remained constant until the hydrometeors were between the +2° and +8° C level when the charge became abruptly negative. This change in the sign of the charge was found to correlate with the final melting of the hydrometeors and MacCready and Proudfit concluded that a strong charging mechanism is associated with the melting of hydrometeors. It should be noted that these results give a sign reversal which is opposite to that found by previous workers.

Latham and Stow (1965 C) showed that the steady reduction of the positive charge at temperatures below 0° C could be explained in terms of charge generation due to the evaporation of the ice particles.

MacCready and Proudfit (1965 B) performed laboratory experiments on the charge associated with melting for comparison with their field measurements. Spheres and cubes of ice were melted in an airstream and the charge measured in three different ways. They found certain consistent features of the charging. The sample always acquired a positive charge of the order of 0.1 e.s.u. and the acquisition of the charge occurred mainly during the later stages of the melting. Results on natural hail samples were found to be generally similar though more erratic. MacCready and Proudfit concluded that their results supported the observations of Dinger and Gunn although they did not find the charging to be as sensitive to impurities as Dinger and Gunn had found. They also found that the rate of melting of the samples affected the magnitude of the charge and suggested that this might be the cause of the difference in the results of Dinger and Gunn, and of Mathews and Mason. They further suggested that the difference between their field and laboratory measurements might be due to the different ambient conditions.

3. ELECTRIFICATION ASSOCIATED WITH TEMPERATURE GRADIENTS IN ICE.

Findeisen (1940) grew a fine rimed deposit on an iced surface maintained at -60° C and observed that the deposit acquired a charge. He suggested that the sign of the charge was determined by the direction of temperature gradient in the ice surface and that a compensating charge was carried away on small ice splinters which were ejected from the surface. In a later investigation (1943) he showed that the splinters did carry this charge.

Reynolds, Brook and Gourley (1957) investigated the electrification resulting from the asymmetric rubbing of ice-coated rods. When the two rods were both coated with pure ice the warmer one was found to acquire a negative charge. However, when one of the specimens was made of a 10^{-4} N solution of sodium chloride it became negative although it was as much as 25° C colder than the specimen of pure ice. They attributed this reversal of sign to the formation of a liquid layer due to the rubbing, followed by selective ion absorption on refreezing of the layer.

Brook (1958) measured the electrification resulting from the transient contact of pieces of ice under conditions when rubbing and frictional contact were minimised. He found the sign of the charge to be related to that of the temperature gradient in the same manner as found by Reynolds, Brook and Gourley although the magnitude of the charge was of an order of magnitude less. Brook suggested that as the conductivity of ice increased with increasing temperature and increasing contamination suggesting a temperature-controlled rate-process, then the conduction process could be described in terms of an effective proton-gas temperature. Two samples of ice in contact at different temperatures would then give rise to an e.m.f. which would cause the transfer of protons from the warm to the cold ice. As this mechanism did not fully explain the results obtained with salty ice he also suggested the existence of a "second-order pyroelectric effect."

Latham and Mason (1961 A) put forward a formal theory of the charge separation due to temperature gradients as described in chapter 2. They confirmed the theoretical results experimentally.

There have been a number of recent publications verifying the existence of a charge separation effect due to temperature gradients. The most convincing experiment is probably that of Latham (1964) in which charge separation was observed in an ice crystal which was in contact solely with a suspending fibre, spurious effects thus being eliminated as far as possible.

4. ELECTRIFICATION ASSOCIATED WITH HAIL

(a) Collisions between ice crystals and hailstones.

Reynolds, Brook and Gourley (1957) investigated the charging produced when an ice-coated sphere moved through a cloud of supercooled water drops and ice crystals. The experiment was performed in a large cold chamber which was maintained at -25° C. A water vapour source was present in the chamber and produced a cloud of supercooled drops of about 5μ diameter. This cloud could be seeded in several ways to produce ice crystals of up to 100μ in size. Two 4 mm diameter ice coated spheres were mounted on the ends of a rotating rod and connected to an electrometer. The spheres were moved through the cloud of drops and ice crystals at a speed similar to that of hailstones of the same size falling through the atmosphere, and the rate of charging observed. The relative concentrations of drops and ice crystals in the cloud was varied and it was found that little or no charging occurred when the cloud was composed entirely of drops or crystals. Positive charging was found to be associated with a high concentration of crystals compared to drops, and negative charging with a high concentration of drops compared to crystals. When the cloud conditions were set to favour positive charging, negative charging could be obtained by warming the ice-coated spheres or by adding sodium chloride smoke to the cloud. Reynolds, Brook and Gourley explained the negative charging as being due to a temperature difference effect in which the spheres were warmed by the latent heat released from the drops freezing onto them and became negatively charged by the collisions of the colder ice crystals. They measured a charging rate of 2 e.s.u. sec^{-1} for a crystal concentration of 10^7 m^{-3} and thus obtained an estimate of 5×10^{-4} e.s.u. for the charge separated per crystal collision. This estimate formed the basis of the thunderstorm theory described in Chapter 1.

Latham and Mason (1961 B) investigated the electrification of an iced probe by ice crystal collisions in the absence of liquid water. The apparatus consisted of an ice-coated insulated rod which was connected to an electrometer. Ice crystals were drawn past the probe in an air stream. The temperatures of the ice surface and of the crystals were varied, and so was the speed at which the crystals were drawn past the probe. The size of the crystals and the number impacting on the rod were measured by making plastic castings on a Formvar-coated rod of similar diameter. Although the crystals were not of uniform size it was found possible to vary their averagesize between 20 and 50μ. The rate of charging of the ice surface was found to be dependent on the temperature difference between the ice crystals and the surface. Variation of the impact velocity of the crystal over the range 1 to 30 m $\sec^{-1}$ produced no systematic variation in the charging. For a temperature difference of 5° C they found that the average charge separated per collision for 20μ size crystals was 5×10^{-9} e.s.u., which is five orders of magnitude less than the value found by Reynolds, Brook and Gourley. When the ice coating on the rod was contaminated with 3.6 mg ℓ^{-1} of sodium chloride, which is the concentration normally found in clouds, the effect on the charging was equivalent to raising the temperature of the ice surface by 2° C.

Magono and Takahashi (1963) passed a stream of ice crystals of about 0.5 mm diameter past an iced probe. The temperatures of the probe and the crystals could be varied. Under these conditions no considerable charging was observed. They then passed both ice crystals and drops of about 5μ diameter past the probe and observed the charging. Their results agreed on the whole with those of Reynolds, Brook and Gourley. Magono and Takahashi then investigated the effect of the nature of the ice surface on the charging and found that the probe became negatively charged by collision with ice crystals if the surface was freshly rimed. They found that this effect could enhance the charging by up to a factor of 6 as compared with a glazed surface. However, it is not certain that these results are applicable to the smaller ice crystals present in thunderclouds.

Church (1966) investigated the electrification due to ice crystal collisions by drawing supercooled drops and ice crystals past a rotating probe which consisted of 4 ice-coated spheres of 4 mm diameter on opposite ends of two rods which were connected to an electrometer. The results were generally similar to those of Reynolds, Brook and Gourley except that the charge separated per crystal collision was found to be 10^{-6} e.s.u.

(b) Collisions between supercooled water drops and hailstones

Several workers have measured the charge produced when supercooled water drops encounter an ice surface, and have generally found that the ice surface acquired a negative charge.

As an exception to this Findeisen (1940) found that the ice surface acquired a positive charge as soon as the drops began to freeze. The charge was reduced if the ice surface became smooth and glassy, or wet which happened when the drops froze slowly. A natural supercooled cloud gave rather larger charging than an artificial spray and Findeisen suggested that this was due to the more rapid freezing of the smaller cloud drops. Kramer (1948) repeated Findeisen's work in greater detail and found that the ice surface acquired a negative charge which was proportional to the impact velocity of the drops. Lueder (1951A,B) performed experiments in natural supercooled clouds on a mountain top and found that the growing rime deposit acquired a negative charge, positive charge presumably being carried by the parts of the drops which were flung off without freezing. Meinhold (1951) measured the electric field strength at the surface of an aircraft flying through a supercooled cumulus congestus cloud and thus found the rate of charging of the aircraft due to riming. The aircraft acquired a negative charge at a rate of 5×10^{-12} C cm^{-2} sec^{-1}. Weickmann and aufm Kampe (1950) sprayed supercooled water drops of between 5 and 100 μ diameter onto a 5 mm diameter metal rod. They found the charging to be insensitive to dissolved salts and proportional to the impact velocity of the drops. At a velocity of 15 m sec^{-1} the charging rate attained a value of 5×10^{-12} C cm^{-2} sec^{-1} in good agreement with Meinhold's results. However, they later suggested that the charging might have been seriously affected by electrification associated with the production of the spray. Reynolds, Brook and Gourley (1957) found that charging occurred only in the presence of ice crystals. Latham and Mason (1961 B) measured the electrification by drawing supercooled drops in the diameter range 40 to 100 μ in an air stream past a stationary ice-coated sphere which was connected to an electrometer. For impact velocities between 5 and 15 m sec^{-1} the drops were found to produce ice splinters when they impinged on the probe. The probe acquired a negative charge which was proportional to the number of splinters produced. On average Latham and Mason found that each drop produced 12 splinters and caused a charge separation of 4×10^{-6} e.s.u. This formed the basis of the theory of thunderstorm electrification discussed in Chapter 1. Magono and Takahashi (1963) found that charging was produced only in the presence of ice crystals. They also found that the sign of the charge was dependent on the temperature of the probe and the riming rate. Church (1966) found that riming by supercooled water drops produced no charge unless ice crystals were present or the drops were freezing when they impinged on the probe.

5. ELECTRIFICATION BY THE EVAPORATION OF ICE.

As stated in Chapter 2 an evaporating ice specimen will develop a temperature gradient across its surface and this will lead to the specimen becoming negatively charged as the outer surface is stripped off. Latham and Stow (1965 A) measured the electric current produced when an ice-coated copper sphere of diameter 3.4 cm was exposed to a stream of chilled nitrogen. The sphere was maintained at a temperature of -20° C while the temperature of the nitrogen was varied between 0° C and -40° C. For nitrogen temperatures above -10° C the sphere became positively charged while below -10° C it was always negatively charged. In a separate experiment the variation of the temperature gradient in the ice surface with the temperature of the nitrogen was found. Using these results they showed that the current due to the evaporation increased smoothly with the temperature gradient in the ice surface. The maximum rate of charging of the sphere was found to be 4×10^{-4} e.s.u. sec^{-1}. For a hailstone of diameter 4 mm the rate of charging would be about two orders of magnitude less. If hailstones are the generators of the electrification of thunderclouds they must on average each be charging at a rate of at least 10^{-3} e.s.u. sec^{-1}, which is greater than the charging produced by evaporation. While this charging is therefore not sufficient to account for thunderstorm electrification it may be a source of positive space charge in dry air blowing over snow, or in dry air through which snow and hail is falling.

6. OTHER SOURCES OF ELECTRIFICATION.

The Workman - Reynolds effect has been described in Chapters 1 and 2. As stated then it is sensitive to the presence of impurities and no charging has been observed due to the impaction of drops on a wet surface by other workers.

The Wilson process of selective ion capture is probably of importance in determing the sign of charge on precipitation reaching the ground.

7. FIELD MEASUREMENTS.

Field measurements of the charge on hail will only be of significance to thunderstorm electrification if the conditions in the cloud from which the hail has fallen are known. Usually these conditions are not known. The discrepancies between the few results which have been obtained are probably due to the different conditions under which observations were made.

Küttner (1950) measured the charge on hail inside clouds on the Zugspitze and found it to be nearly always positive. Moore (1965) found that the charge on hail collected at the ground varied from minute to minute and was usually of the same sign as the point discharge ions being produced by the high potential gradient. MacCready and Proudfit (1965) measured the charge on individual hailstones in and below thunderclouds. At temperatures well below 0° C they observed charges of a few e.s.u., while at temperatures near 0° C the hailstones had a smaller positive charge. When the hailstones melted the charge became negative. The positive charges are consistent with the results of Küttner. Moore's results may be interpreted in terms of the Wilson selective ion capture process, but it is not known whether this had any effects on the results of Küttner, and of MacCready and Proudfit.

8. SUMMARY.

The laboratory experiments described in this chapter are open to doubt as far as their applicability to natural conditions is concerned. In all cases the airflow around the ice specimens, and thus the rate of heat transfer, is probably unlike that occurring in natural conditions. Also, in most of the experiments the charge was measured by being leaked away to earth through an electrometer and thus the effect of the building up of charge on the specimens was not found. The experiments of Magono and Kikuchi (1963, 1965), and of Kikuchi (1965) most closely approach natural conditions. However, their results are of doubtful validity insofar as the snowflakes were melted rapidly while MacCready and Proudfit found that the rate of melting affected the electrification produced.

For these reasons it was decided to investigate the electrification of ice specimens which were freely supported in a stream of air.

CHAPTER 7.

APPARATUS FOR FREELY SUSPENDING ICE.

1. AIR NOZZLE.

It was found that solid bodies could be suspended by the airflow issuing from a nozzle even when their shape departed considerably from spherical. The most consistent results were obtained when the nozzle diameter was less than a quarter of the diameter of the object being supported. Although there was found to be a lower limit to the nozzle diameter for any particular size of body it was wished to support, the actual size of nozzle used was not found to be critical over a wide range. Attempts were made to support bodies of from 4 mm to 3 cm in diameter and it was found that bodies over 6 or 7 mm could be stably supported on a nozzle of diameter 3/32". Larger bodies were supported on a nozzle of diameter 1/4". It was found that this means of support was so stable that bodies could be supported by the airflow at angles of up to 45° to the vertical. When 4 mm diameter ball bearings were placed in the airstream from a 1/8" diameter nozzle they were observed to be supported for a period of 1 or 2 seconds before becoming unstable. It seems likely that it would be possible to support small objects of this size by using a smaller nozzle; unfortunately there was not sufficient time available to verify this.

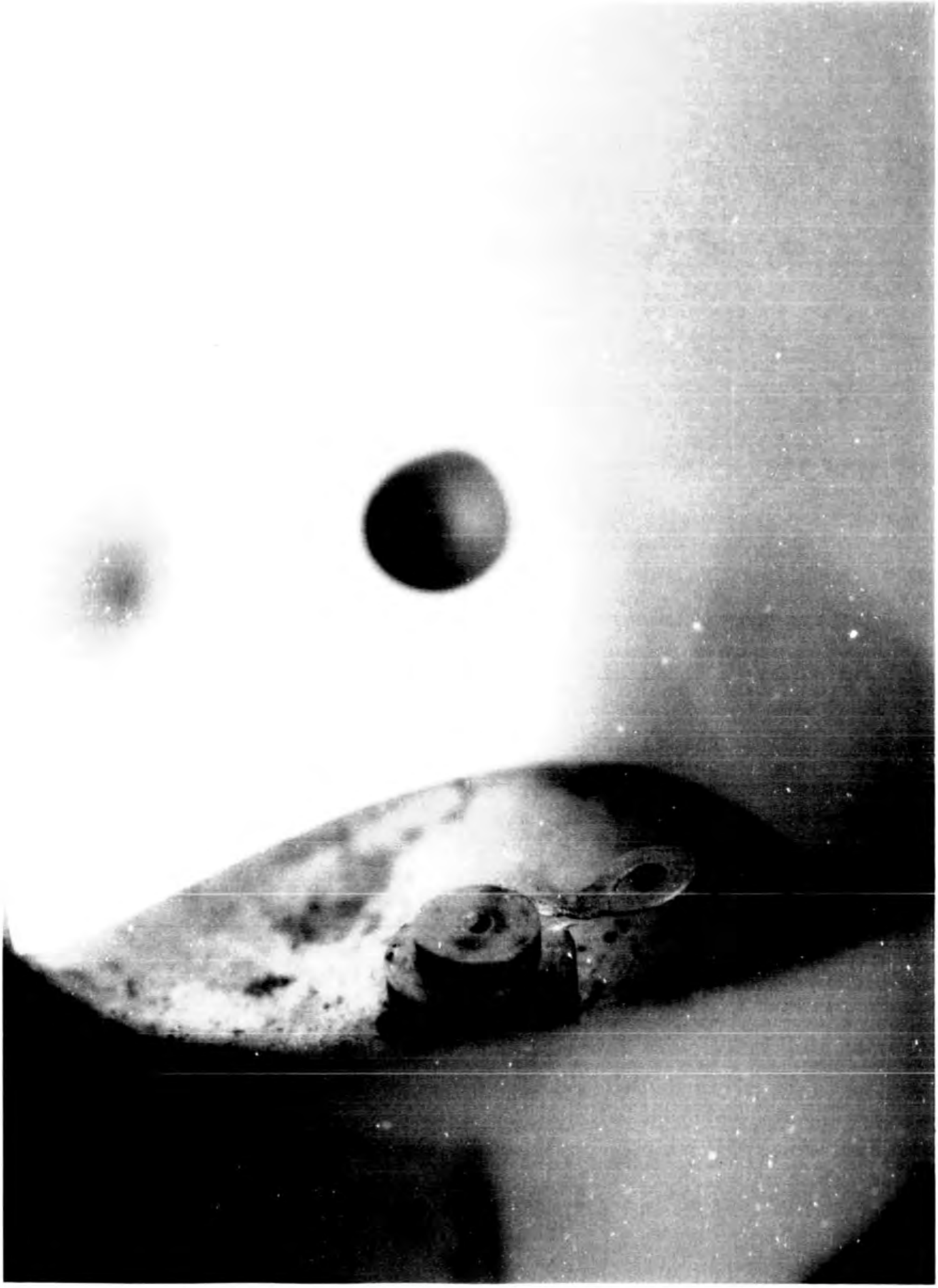


Fig. 14. The Air Nozzle Supporting a Plasticine Sphere.

Accordingly a nozzle of diameter 3/32" in a 1/2" diameter brass rod was used to support spheres of ice of about 15 mm diameter.

2. THE ICE MOULDS.

While it was not necessary for the ice particles to be spherical in order to be supported in the airflow it was decided that measurement would more conveniently be made on spherical particles of uniform size. It was found that hollow glass balls of the type used for Christmas decorations made satisfactory moulds. They could be easily obtained and varied in diameter by only 1 mm either side of 15 mm. To make the ice spheres the balls were filled with water through the hole already in them and then placed in a refrigerator. After the water had frozen the glass shell was broken away. Care was taken to make sure that all the glass was removed from the ice sphere.

3. THE CHARGE COLLECTOR.

Initially measurements on the charge acquired by the ice spheres after a period of suspension in the supporting air stream were made by blowing the sphere into a cylindrical charge collector which was connected to the electrometer used in the previous investigation. The sphere was blown out of the supporting air stream by a short burst of air from a secondary nozzle. This worked satisfactorily except that considerable skill was required to blow the sphere into the charge collector, and on average only half of the spheres were caught. Because of this the charge collector was changed. The air nozzle was mounted in the base of a cylindrical can of diameter 6.5 cm and height 3 cm and was electrically connected to it. The charge collector and the nozzle were insulated from earth and connected to the input of the electrometer. The charge on the spheres was measured by cutting off the air supply and allowing the sphere to fall back into the charge collector. The charge collector and nozzle were mounted on an earthed metal plate inside a refrigerated cell. The walls of the cell were of stainless steel and were earthed so as to shield the charge collector from extraneous fields. The electrometer was used with the $10^{10}\,\Omega$ input resistor selected and the indicator unit set for 1000 mV full scale deflection. Tests showed the noise level of the charge collector system to be of the order of 50 mV under these conditions of operation. This instrument noise set the minimum charge measurable as 15×10^{-3} e.s.u. The output from the electrometer was connected to the Watanabe pen recorder.

4. THE AIR SUPPLY AND COOLING SYSTEMS.

The air supply and cooling apparatus were as described in Chapter 4 except that the length of capillary tubing was removed from the air supply line. The temperature of the air stream at the outlet nozzle was measured by means of a thermocouple and it was found that the lowest temperature attainable was - 14° C. It was also found that large numbers of small water drops were produced in the air flow and these were utilised in the investigation into the electrification associated with the riming of the ice spheres.

The general layout of the apparatus was similar to that described in Chapter 4 except that the air nozzle and charge collector replaced the support tube.

CHAPTER 8.

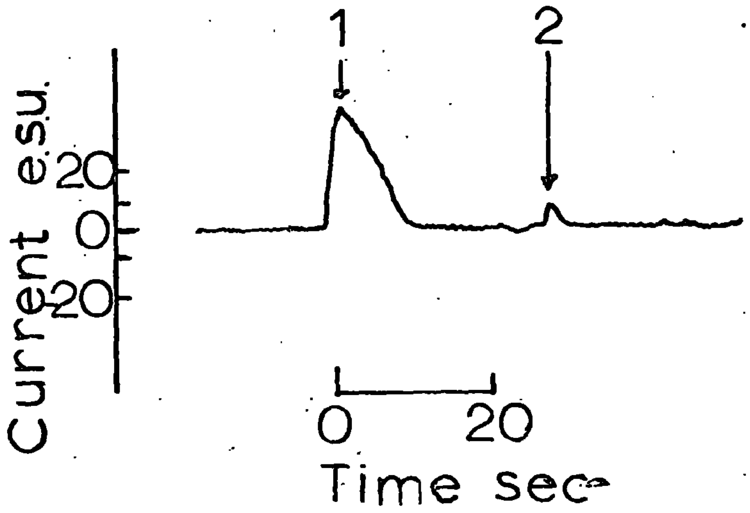
RESULTS.

1. EXPERIMENTAL METHOD.

Measurements were made of the electrification associated with the riming and melting of the ice spheres. The experimental method was similar in both investigations. The air flow was set to the value required to support the ice sphere about 8 cm above the charge collector as found by experiment. Then the temperature of the airflow was adjusted to the required value and allowed to stabilise. The electrometer input selector was set on the "short circuit" input and then an ice sphere was introduced into the airflow, metal forceps being used to handle it. The $10^{10}\,\Omega$ input resistor was selected on the electrometer. After the sphere had been supported for a known length of time, usually about 20 sec, the air supply was cut off and the sphere allowed to fall back into the charge collector,

2. ELECTRIFICATION ASSOCIATED WITH RIMING OF THE ICE SPHERES

Measurements were made of the electrification due to riming of the ice spheres of diameter 15mm at air temperatures -7° C, -10° C, -12° C and -13° C. The spheres were rimed by accretion of the droplets already present in the supporting air stream as mentioned in Chapter 7. The size of the droplets was found by allowing them to impinge on a Formvar-coated glass plate. The size of the droplets was found to vary between 5 and 100μ with the mean value being about 25μ. The number of drops impinging on the plate could only be found very approximately but appeared to be of the order of 10^4 sec^{-1}. No ice crystals were observed in the air stream but it is not certain that there were none present as crystals smaller than 5μ would not have been detected. In order to find the percentage of drops present in the airstream which collided with the ice sphere being suspended, droplets which had been coloured with potassium permangate were sprayed into the air stream. Those droplets which did not collide with the ice sphere were collected on a piece of blotting paper which was situated in the air stream above the sphere. Tests in the absence of any ice sphere showed this to be a satisfactory means of collecting the coloured droplets. When an ice sphere was being supported in the air stream no discolouration of the blotting paper was observed indicating that all the droplets were colliding with the sphere. Measurements were made under otherwise similar conditions both on the same sphere repeatedly, and on a series of freshly made spheres, in order to see if any effect due to ageing of the sphere surface could be detected. No



1. 10^{10} Ω Input Selected
2. Charge on Sphere.

Fig. 15. Recorder Trace of a Charge Measurement on a Riming Ice Sphere.

such effect was found, but it was decided to use a freshly made sphere for each measurement so as to eliminate this effect if it existed. In order to make the measurements as nearly identical as possible each sphere was heated by means of the radiation from an electric light bulb until the surface was covered by a thin layer of liquid water, and then introduced into the air stream. The layer of liquid quickly froze when the sphere was supported in the air stream.

The results of the measurements are shown in Table 5 and a typical recorder trace is shown in Fig.15.

TABLE. 5. Electrification of 15 mm diameter ice spheres after riming for 20 sec.

No.	Air temperature °C	Charge e.s.u.
1	- 7	-15
2	- 7	- 3
3	- 7	-12
4	- 7	-14
5	-10	-12
6	-10	- 6
7	-10	-12
8	-10	-14
9	-10	-15

No.	Air temperature °C	Charge e.s.u.
10	-10	-18
11	-10	- 4
12	-10	- 6
13	-10	-10
14	-10	-13
15	-12	+12
16	-12	+ 9
17	-12	+10
18	-12	+12
19	-12	+ 3
20	-12	+ 7
21	-12	+39
22	-13	+ 3
23	-13	+ 7
24	-13	+ 5

The large charge of +39 e.s.u. measured on sphere No.21 is thought to be spurious as this sphere was observed to rime considerably more than the others. It is thought that this was due to the presence of a cloud of droplets in the refrigerated cell which was produced by the operator's breath. Although an insufficient number of results was obtained to allow the drawing of any conclusions as to the variation in magnitude of the electrification with temperature, there appears to be a definite change in the sign of the charge at between -10° and -12° C. To find whether this sign reversal was dependent on the air temperature or the temperature of the spheres, measurements were made on spheres which were at different temperatures when introduced into the air stream. The sign reversal was still found to take place at about -10° C indicating that the temperature of the air stream is the determining factor.

A possible explanation of this sign reversal is based on the observation of the change in the freezing behaviour of single drops reported in Chapter 5. As the droplets which rimed the spheres were produced by condensation in the air stream, and being small would have short thermal relaxation-times, it is reasonable to expect that they would be at the temperature of the air stream when they collided with the ice spheres. Drops which were nucleated at temperatures above -10° C by collision with the ice spheres would be expected to freeze slowly from the point of impact and some of the liquid portion of the drop would splash off the sphere and be carried away in the airstream. Charge could thus be separated either by the effect of the splashing or by the development of a Workman-Reynolds potential across the freezing face of the drops. Drops which collided with the sphere at temperatures below -10° C would be expected to freeze to form a covering ice shell. Further freezing of the drops would then lead to their breaking and ejecting charged ice splinters.

While this explanation appears to be feasible it raises several questions. Magono and Takahashi (1963) found that the sign of the charge acquired by a riming iced probe depended on the temperature of the probe and the riming rate. They found that for a rate of riming equal to that of the hailstone in a thundercloud the sign of the charge acquired by the hailstone would change from negative to positive as the hailstone fell past the -10° C level. This sign reversal is of opposite polarity to that found in this investigation. The explanation of this effect given by Magono and Takahashi depends on collisions between ice crystals and the probe, the riming due to supercooled drops being of importance only in determining the nature of the probe surface. The difference in polarity of the sign reversal and the apparent absence of ice crystals in the present investigation indicate that different charge generating mechanisms were operating in the two cases.

The negative charge acquired by the ice spheres when rimed by droplets at temperatures above -10° C agrees with the findings of previous workers as to the charge acquired by a riming surface, except for the results of Findeisen. The positive charge acquired when the spheres were rimed by droplets at temperatures below -10° C is of opposite sign to that observed by most previous workers and it is difficult to explain the origin of this charge.

While the accretion rate was not accurately measured it is of interest to calculate an approximate value of the charge separated per droplet collision. As already stated nearly all of the droplets present in the air stream collided with the ice spheres. Thus, if there are assumed to be 10^4 collisions per sec as stated in Chapter 7, after 20 sec there would have been 2×10^5 collisions. Taking 10 e.s.u. as the charge acquired by the ice sphere in this time the average charge per collision is 5×10^{-5} e.s.u. This is of the same order of magnitude as the values found by Reynolds, Brook and Gourley (1957), and by Church (1966).

Because of the small number of results and the uncertainty in the rate of accretion care must be taken in applying these results to the electrification of thunderclouds. However, if a hailstone falling in a thundercloud were accreting droplets in a similar manner, the results indicate that at levels above the -10° C isotherm it would acquire a positive charge by riming. The negatively charged particles flung off would be carried up towards the top of the cloud in the updraught. When the hailstone fell below the -10° C level, riming would cause it to acquire negative charge and its net charge would eventually become negative. Again, the positively charged particles flung off would be carried up to the top of the cloud in the updraught. If the charges carried to the top of the cloud are of equal magnitude for both signs of charge then this would give rise to a positive charge above the -10° C level and a negative one beneath it. The measurements of MacCready and Proudfit have shown that the charge carried by hailstones at the -8° C level are generally positive. If some mechanism operates at the -8° C level which reverses the polarity of the charge on the hailstones then the above theory would account for the concentration of the negative charge near the -10° C level.

3. ELECTRIFICATION ASSOCIATED WITH THE MELTING OF ICE.

Measurements were made of the charge acquired by ice spheres of diameter 15 mm when they melted in an air stream at a temperature of 18° C. The spheres were supported in the air stream for a known length of time and their behaviour during melting observed. The degree of melting was not measured except as to whether or not it had progressed to the point where visible drops of water were being flung off. The results of the experiment are shown in Table 6.

TABLE 6. Electrification associated with melting of ice spheres of diameter 15 mm.

No.	Charge e.s.u.	Degree of melting.
1	+ 5	1
2	+ 6	1
3	+ 8	1
4	- 6	2
5	- 9	2
6	- 4	2
7	- 3	2

The figures 1 and 2 in the third column of Table 6 indicate:

1) that the sphere was covered by a film of liquid water but no visible drops had been flung off.

2) that visible drops of water had been flung off from the sphere.

Although the number of results is too small to allow of definite conclusions being drawn they indicate that the charge on the spheres depends on whether or not water had been thrown off. The spheres were opaque and apparently contained large numbers of air bubbles. A possible explanation of the results is that so long as no water was flung off the charge mechanism was that of air bubble release as observed by previous workers. When water was flung off the dominant charge mechanism was that of the breaking of the water surface. This is consistent with the proposed explanation of the charging due to riming at drop temperatures above -10° C.

Again caution must be observed in applying these results to thundercloud electrification, both because of the small number of results and because of the large size of the ice spheres. The maximum stable size of raindrop in the atmosphere is about 5 mm. In the turbulent conditions of a thundercloud this limiting size may well be less. Also, as described in Section 4 of this chapter, melting ice spheres were observed to rotate rapidly about a vertical axis after the formation of a liquid film on their surface. If this is true of hailstones then the disruptive forces due to this spinning may lower the maximum stable size of raindrop. If these factors reduce the maximum stable size of raindrop to about 3 mm then an explanation may be found for the results of MacCready and Proudfit. In their laboratory experiment no water would be flung off from the ice sample and thus the charge acquired by the sample would be positive as they found.

In their field measurement they observed the charge on melting hailstones of about 4 mm diameter. If the raindrops resulting from the melting of the hailstones are taken to be unstable for sizes above 4 mm diameter then it would be expected that the melting hailstone would be stable until the completion of melting. If this is the case then until melting was complete no water would be flung off and the charge on the hailstone would be positive. On the completion of melting the raindrop would be unstable and water would be flung off; thus the raindrop would acquire a negative charge. This is consistent with the observations of MacCready and Proudfit.

4. THE MELTING BEHAVIOUR OF THE ICE SPHERES.

The melting behaviour of spheres of diameter 15 mm and 40 cm was observed. It was similar for both sizes of sphere. As long as the surface of the spheres was dry they remained stationary in the air stream without rotating. Melting commenced by the growth of a film of water which spread from the base of the sphere over its entire surface. As the melting progressed further the sphere began to rotate rapidly about a vertical axis. The rate of rotation was measured with a stroboscope and found to be of the order of 300 to 400 revolutions per minute. Further melting of the sphere led to the formation of a ring of water about the horizontal equator of the sphere. Drops of water of about 2 mm diameter were then flung off from this ring and carried away in the air stream. This behaviour is similar to that observed by Blanchard (1953) for the melting of ice specimens suspended in an air stream by means of fibres. Blanchard did not observe the spinning of the specimens but this was probably due to the effect of the suspending fibre. He observed that the drops flung off from the ring of water passed over the top of the specimen before being carried away in the air stream. This was occasionally observed in the present experiment but usually the drops did not do this. This is probably due to the spinning motion of the spheres which tended to fling the drops off in a nearly horizontal direction.

CHAPTER 9.

CONCLUSIONS AND SUGGESTIONS FOR FURTHER WORK.

Experiments on the freezing and electrification of individual water drops suspended by an air stream have shown considerable differences in the behaviour of these drops as compared with drops which were supported on a fibre in stagnant air conditions. The freezing behaviour of the drops has been shown to be temperature-dependent, a marked change in behaviour occurring for temperatures above and below -10° C. The experiments have also shown that no electrification was produced by the freezing of these drops within the experimental limit of $\pm 0.15 \times 10^{-3}$ e.s.u. As these observations were made under conditions more closely resembling the conditions in a thundercloud than previously they throw doubt on the applicability of previous results to thunderstorm electrification. These discrepancies also raise the question as to how far the present laboratory results may be applied to electrification in thunderclouds.

Experiments on the electrification of ice spheres supported in an air stream have shown the existence of what would appear to be a temperature-dependent mechanism of charging due to riming. Riming by the accretion of droplets warmer than -10° C leads to the acquisition of a negative charge by the ice spheres, while riming by the accretion of droplets colder than -10° C leads to the acquisition of a positive charge. It is suggested than an explanation of this effect lies in the temperature - dependence of the freezing behaviour of drops mentioned above. It is also suggested that this mechanism could give rise to thundercloud electrification of the observed polarity, and also the observed concentration of negative charge near the -10° C level. The experiments have also shown that the electrification of a melting hailstone appears to depend on whether or not water is flung off during melting. If water is not flung off then the hailstone acquires a positive charge, if water is flung off the hailstone becomes negatively charged and it is thought that this can explain the difference between the field and laboratory measurements of MacCready and Proudfit.

The melting behaviour of ice supported by an air stream has been shown to be as found by Blanchard except that the ice spheres were observed to rotate rapidly about a vertical axis when their surfaces became wet. It is proposed that this would reduce the maximum size of stable raindrop resulting from the melting of hailstones.

1. SUGGESTIONS FOR FURTHER WORK.

The differences in the results of the present investigations as compared with previous experiments under less natural conditions of support throw doubt on the applicability of laboratory results to thundercloud electrification. The results underline the need in future work to have the experimental conditions more nearly resemble those in the atmosphere.

APPENDIX I

OBSERVATIONS.

No.	Drop diameter mm	Air temperature °C	Time before freezing sec	Freezing behaviour
1	3.0	- 8.6	16	2
2	3.0	-17.1	12	2
3	3.0	-17.1	14	2
4	3.0	-19.0	12	2
5	3.5	- 4.8	15	1
6	3.5	- 4.8	17	1
7	3.5	- 7.6	15	1
8	3.5	- 7.6	17	1
9	3.5	- 8.6	12	2
10	3.5	- 8.6	14	2
11	3.5	- 8.6	15	2
12	3.5	-10.5	10	2
13	3.5	-10.5	21	2
14	3.5	-13.2	14	2
15	3.5	-14.4	14	2
16	3.5	-17.1	12	2
17	4.0	- 4.8	22	1
18	4.0	- 4.8	25	1
19	4.0	- 5.8	16	1
20	4.0	- 5.8	16	1

No.	Drop diameter mm	Air temperature °C	Time before freezing sec	Freezing behaviour
21	4.0	- 7.6	12	1
22	4.0	- 7.6	18	1
23	4.0	- 8.6	18	2
24	4.0	-10.5	15	2
25	4.0	-13.2	13	2
26	4.0	-13.2	15	2
27	4.0	-14.4	12	2
28	4.0	-14.4	16	2
29	4.0	-17.1	12	2
30	4.0	-17.1	12	2
31	4.0	-17.1	14	2
32	4.0	-19.0	10	2
33	4.0	-19.0	13	2
34	4.5	- 4.8	17	1
35	4.5	- 5.8	16	1
36	4.5	- 5.8	18	1
37	4.5	- 8.6	15	1
38	4.5	- 8.6	20	2
39	4.5	- 8.6	21	2
40	4.5	-10.5	13	2
41	4.5	-13.2	12	2
42	4.5	-19.0	10	2

No.	Drop diameter mm	Air temperature ^{o}C	Time before freezing sec	Freezing behaviour
43	4.5	-19.0	14	2
44	5.0	- 5.8	23	1
45	5.0	- 8.6	18	2
46	5.0	-10.5	15	2
47	5.0	-13.2	14	2
48	5.0	-17.6	12	2
49	5.0	-17.6	12	2
50	5.0	-17.6	11	2
51	3.0	- 5.3	14	1
52	3.0	- 5.3	15	1
53	3.0	- 5.3	19	1
54	3.0	-10.7	16	2
55	3.0	-13.5	15	2
56	3.0	-13.5	17	2
57	3.5	- 5.3	13	1
58	3.5	- 7.9	13	1
59	3.5	- 7.9	16	2
60	3.5	- 9.3	16	2
61	3.5	-13.5	14	2
62	3.5	-17.6	13	2
63	4.0	- 5.3	15	1
64	4.0	- 7.9	14	1
65	4.0	- 7.9	14	1

No.	Drop diameter mm	Air temperature ^{o}C	Time before freezing sec	Freezing behaviour
66	4.0	- 7.9	15	2
67	4.0	- 8.2	13	1
68	4.0	- 8.2	15	2
69	4.0	- 8.2	15	2
70	4.0	- 9.3	12	1
71	4.0	- 9.3	14	1
72	4.0	- 9.3	14	2
73	4.0	- 9.3	17	2
74	4.0	-10.7	14	2
75	4.0	-10.7	15	2
76	4.0	-13.5	12	2
77	4.0	-13.5	12	2
78	4.0	-13.5	13	2
79	4.0	-13.5	16	2
80	4.0	-15.2	11	2
81	4.0	-15.2	12	2
82	4.0	-17.6	10	2
83	4.0	-17.6	10	2
84	4.0	-17.6	12	2
85	4.0	-19.3	13	2
86	4.0	-19.3	13	2
87	4.5	- 5.3	14	1
88	4.5	- 5.3	17	1

No.	Drop diameter mm	Air temperature ^{o}C	Time before freezing sec	Freezing behaviour
89	4.5	- 7.9	15	1
90	4.5	- 7.9	15	1
91	4.5	- 9.3	14	2
92	4.5	-10.7	12	2
93	4.5	-15.2	12	2
94	4.5	-17.6	11	2
95	5.0	- 5.3	19	1
96	5.0	- 7.9	15	1
97	5.0	- 7.9	17	1
98	5.0	- 7.9	17	1
99	5.0	-13.5	14	2
100	5.0	-19.3	13	2
101	3.0	- 4.7	19	1
102	3.0	- 4.7	15	1
103	3.0	- 7.3	16	2
104	3.0	- 9.6	13	2
105	3.0	- 9.6	13	2
106	3.0	-13.1	12	2
107	3.0	-13.1	14	2
108	3.5	- 4.7	14	1
109	3.5	- 4.7	17	1
110	3.5	- 5.4	15	1

No.	Drop diameter mm	Air temperature °C	Time before freezing sec	Freezing behaviour
111	3.5	- 5.4	15	1
112	3.5	- 7.3	12	1
113	3.5	- 7.3	13	1
114	3.5	- 7.3	16	2
115	3.5	- 9.6	14	2
116	3.5	-13.1	14	2
117	3.5	-18.7	12	2
118	4.0	- 5.4	17	1
119	4.0	- 5.4	17	1
120	4.0	- 7.3	13	1
121	4.0	- 7.3	14	2
122	4.0	- 7.3	16	2
123	4.0	- 9.6	11	2
124	4.0	- 9.6	11	2
125	4.0	- 9.6	13	2
126	4.0	-13.1	12	2
127	4.0	-13.1	14	2
128	4.0	-15.1	10	2
129	4.0	-15.1	10	2
130	4.0	-15.1	11	2
131	4.0	-15.1	13	2
132	4.0	-18.7	9	2
133	4.0	-18.7	10	2

No.	Drop diameter mm	Air temperature °C	Time before freezing sec	Freezing behaviour
134	4.0	-18.7	10	2
135	4.0	-18.7	11	2
136	4.0	-18.7	11	2
137	4.5	- 4.7	14	1
138	4.5	- 4.7	16	1
139	4.5	- 7.3	14	1
140	4.5	- 9.6	13	2
141	4.5	- 9.6	15	2
142	4.5	-13.1	13	2
143	4.5	-15.1	11	2
144	4.5	-15.1	11	2
145	4.5	-18.7	9	2
146	4.5	-18.7	10	2
147	5.0	- 7.3	15	1
148	5.0	-13.1	12	2
149	5.0	-13.1	14	2
150	5.0	-18.7	10	2

APPENDIX II

CALCULATED DROP TEMPERATURE ON FREEZING.

No.	Drop diameter mm	Dry-bulb temperature °C	Wet-bulb temperature °C	Drop temperature °C
1	3.0	- 8.6	-11	-11
2	3.0	-17.1	-17	-16
3	3.0	-17.1	-17	-16
4	3.0	-19.0	-19	-18
5	3.5	- 4.8	- 8	- 8
6	3.5	- 4.8	- 8	- 8
7	3.5	- 7.6	-10	- 9
8	3.5	- 7.6	-10	-10
9	3.5	- 8.6	-11	-10
10	3.5	- 8.6	-11	-10
11	3.5	- 8.6	-11	-10
12	3.5	-10.5	-12	-10
13	3.5	-10.5	-12	-12
14	3.5	-13.2	-14	-13
15	3.5	-14.4	-15	-14
16	3.5	-17.1	-17	-16
17	4.0	- 4.8	- 8	- 8
18	4.0	- 4.8	- 8	- 8
19	4.0	- 5.8	- 8	- 8
20	4.0	- 5.8	- 8	- 8

No.	Drop diameter mm	Dry-bulb temperature °C	Wet-bulb temperature °C	Drop temperature °C
21	4.0	- 7.6	-10	- 9
22	4.0	- 7.6	-10	-10
23	4.0	- 8.6	-11	-11
24	4.0	-10.5	-12	-11
25	4.0	-13.2	-14	-13
26	4.0	-13.2	-14	-13
27	4.0	-14.4	-15	-14
28	4.0	-14.4	-15	-14
29	4.0	-17.1	-17	-15
30	4.0	-17.1	-17	-15
31	4.0	-17.1	-17	-16
32	4.0	-19.0	-19	-18
33	4.0	-19.0	-19	-18
34	4.5	- 4.8	- 8	- 8
35	4.5	- 5.8	- 8	- 8
36	4.5	- 5.8	- 8	- 8
37	4.5	- 8.6	-11	-10
38	4.5	- 8.6	-11	-11
39	4.5	- 8.6	-11	-11
40	4.5	-10.5	-12	-11
41	4.5	-13.2	-14	-12
42	4.5	-19.0	-19	-15

No.	Drop diameter mm	Dry-bulb temperature °C	Wet-bulb temperature °C	Drop temperature °C
43	4.5	-19.0	-19	-17
44	5.0	- 5.8	- 8	- 8
45	5.0	- 8.6	-11	-10
46	5.0	-10.5	-12	-11
47	5.0	-13.2	-14	-12
48	5.0	-17.6	-18	-16
49	5.0	-17.6	-18	-16
50	5.0	-19.0	-19	-16
51	3.0	- 5.3	- 8	- 8
52	3.0	- 5.3	- 8	- 8
53	3.0	- 5.3	- 8	- 8
54	3.0	-10.7	-12	-12
55	3.0	-13.5	-14	-14
56	3.0	-13.5	-14	-14
57	3.5	- 5.3	- 8	- 7
58	3.5	- 7.9	-10	- 9
59	3.5	- 7.9	-10	-10
60	3.5	- 9.3	-11	-11
61	3.5	-13.5	-14	-13
62	3.5	-17.6	-18	-17
63	4.0	- 5.3	- 8	- 8
64	4.0	- 7.9	-10	- 9
65	4.0	- 7.9	-10	- 9

No.	Drop diameter mm	Dry-bulb temperature °C	Wet-bulb temperature °C	Drop temperature °C
66	4.0	- 7.9	-10	-10
67	4.0	- 8.2	-10	- 9
68	4.0	- 8.2	-10	-10
69	4.0	- 8.2	-10	-10
70	4.0	- 9.3	-11	-10
71	4.0	- 9.3	-11	-10
72	4.0	- 9.3	-11	-10
73	4.0	- 9.3	-11	-11
74	4.0	-10.7	-12	-11
75	4.0	-10.7	-12	-12
76	4.0	-13.5	-14	-13
77	4.0	-13.5	-14	-13
78	4.0	-13.5	-14	-13
79	4.0	-13.5	-14	-14
80	4.0	-15.2	-16	-14
81	4.0	-15.2	-16	-14
82	4.0	-17.6	-18	-16
83	4.0	-17.6	-18	-16
84	4.0	-17.6	-18	-16
85	4.0	-19.3	-19	-18
86	4.0	-19.3	-19	-18
87	4.5	5.3	- 8	- 7
88	4.5	5.3	- 8	- 8

No.	Drop diameter mm	Dry-bulb temperature °C	Wet-bulb temperature °C	Drop temperature °C
89	4.5	- 7.9	-10	- 9
90	4.5	- 7.9	-10	- 9
91	4.5	- 9.3	-11	-10
92	4.5	-10.7	-12	-11
93	4.5	-15.2	-16	-14
94	4.5	-17.6	-18	-14
95	5.0	- 5.3	- 8	- 8
96	5.0	- 7.9	-10	- 9
97	5.0	- 7.9	-10	- 9
98	5.0	- 7.9	-10	- 9
99	5.0	-13.5	-14	-12
100	5.0	-19.3	-19	-16
101	3.0	- 4.7	- 8	- 8
102	3.0	- 4.7	- 8	- 8
103	3.0	- 7.3	-10	-10
104	3.0	- 9.6	-12	-12
105	3.0	- 9.6	-12	-12
106	3.0	-13.1	-14	-14
107	3.0	-13.1	-14	-14
108	3.5	- 4.7	- 8	- 8
109	3.5	- 4.7	- 8	- 8
110	3.5	- 5.4	- 9	- 9
111	3.5	- 5.4	- 9	- 9

No.	Drop diameter mm	Dry-bulb temperature °C	Wet-bulb temperature °C	Drop temperature °C
112	3.5	- 7.3	-10	- 9
113	3.5	- 7.3	-10	-10
114	3.5	- 7.3	-10	-10
115	3.5	- 9.6	-12	-12
116	3.5	-13.1	-14	-14
117	3.5	-18.7	-19	-17
118	4.0	- 5.4	- 9	- 9
119	4.0	- 5.4	- 9	- 9
120	4.0	- 7.3	-10	- 9
121	4.0	- 7.3	-10	-10
122	4.0	- 7.3	-10	-10
123	4.0	- 9.6	-12	-11
124	4.0	- 9.6	-12	-11
125	4.0	- 9.6	-12	-11
126	4.0	-13.1	-14	-13
127	4.0	-13.1	-14	-13
128	4.0	-15.1	-16	-13
129	4.0	-15.1	-16	-13
130	4.0	-15.1	-16	-13
131	4.0	-15.1	-16	-14
132	4.0	-18.7	-19	-15
133	4.0	-18.7	-19	-15
134	4.0	-18.7	-19	-15

No.	Drop diameter mm	Dry-bulb temperature °C	Wet-bulb temperature °C	Drop temperature °C
135	4.0	-18.7	-19	-16
136	4.0	-18.7	-19	-16
137	4.5	- 4.7	- 8	- 7
138	4.5	- 4.7	- 8	- 8
139	4.5	- 7.3	-10	- 9
140	4.5	- 9.6	-12	-11
141	4.5	- 9.6	-12	-11
142	4.5	-13.1	-14	-12
143	4.5	-15.1	-16	-13
144	4.5	-15.1	-16	-13
145	4.5	-18.7	-19	-15
146	4.5	-18.7	-19	-15
147	5.0	- 7.3	-10	- 9
148	5.0	-13.1	-14	-12
149	5.0	-13.1	-14	-12
150	5.0	-18.7	-19	-15

APPENDIX III

THE TESTING OF THE CHARGE COLLECTOR.

The charge collector described in Chapter 4 was tested by using it to measure known charges on water drops. Charged water drops were produced by applying a potential to a water dropper. The charge on the drops was varied by varying the applied potential, and measured by collecting the drops in a nearly completely closed charge collector. The charge collector described in Chapter 4 was tested twice in this way during the taking of the observations reported in Chapter 5. It was also tested before and after each measurement by bringing a charged body, usually the operator's hand, near to it and observing that the electrometer registered a deflection.

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