A laboratory study of charge transfer accompanying the collision of ice crystals with a simulated hailstone

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SUMMARY

In a series of experiments, conducted inside a cold-room, clouds of small ice crystals of mean diameter \(10 \mu m\) were drawn at velocities between 1.5 and 18 m s\(^{-1}\) past simulated hailstones. Two of these ice targets, side by side but under different conditions, were subjected to crystals from the same clouds simultaneously, so that the effects of various parameters on charge transfer could be isolated. The concentrations, sizes and shapes of the ice crystals were determined.

A target heated radiatively with respect to the crystals became charged more negatively than an unheated target. A colder target became charged more positively. However, the magnitude of the charging did not depend on the temperature difference between target and crystals, provided this was greater than about 1 K. Attempts to find evidence of discharge-limited charging produced negative results. A strong velocity dependence of charging currents was found, but subsidiary experiments on the collection of crystals by the target suggested that this was largely due to the variation in the probability of separation of the crystals, which appeared to be close to zero at the lowest velocities employed. The effects of doping the targets with HF and NH\(_3\), and of surface roughness, were investigated and were found to be small.

These results were found to be in reasonable qualitative and quantitative agreement with the predictions of the charging mechanisms proposed by Auf der Maur and Buser, involving differences in the work functions of ice surfaces of different nature.

It was estimated that the charge, \(q'\), transferred in a rebounding collision between an ice crystal and a heated ice target is about 900 \(\mu C\). It is shown that this is roughly compatible with the general pattern of results of other workers (with one exception) if the charging increases with the square of the diameter, \(d\), of the small particles. There are good theoretical reasons for such a relationship, which may be described by the equation \(q' (\mu C) = 5d^2\), for \(10 \mu m < d < 200 \mu m\). Charge transfer of this magnitude is estimated to be just sufficient to account for the electrification of thunderclouds.

1. INTRODUCTION

Despite intensive research over the past two decades the mechanism or mechanisms responsible for the growth of strong electric fields in thunderstorms, culminating in lightning, remain to be identified. In a recent paper, Illingworth and Latham (1977) concluded from a theoretical assessment of precipitation mechanisms of thundercloud electrification that the two most powerful ones involve collisions between ice particles and hailstones. These are the inductive mechanism, studied by Müller-Hillebrand (1954), Latham and Mason (1962), Sartor (1967), Scott and Levin (1970) and many other workers; and the non-inductive ice–ice process investigated by Reynolds, Brook and Gourley (1957).

However, experimental evidence on both these processes is incomplete and often contradictory. Measurements of the charge transfer accompanying the collision and separation of ice particles in the absence of impressed electric fields have been made by Reynolds, Brook and Gourley (1957), Latham and Mason (1961a), Magino and Takahashi (1963a, b), Latham and Miller (1965), Church (1966), Buser and Auf der Maur (1971), Auf der Maur and Buser (1974) and several other investigators. Values of charge transfer per collision ranging over five orders of magnitude have been obtained. The earlier workers explained the results quantitatively in terms of the thermoelectric effect in ice (Brook 1958; Latham and Mason 1961b; Jaccard 1963; and others), but this process appears to be quantitatively inadequate, and Auf der Maur and Buser offered an interpretation in terms of surface potential differences (Takahashi 1969a, b, 1973; Lowell 1975).
In the hope of resolving some of these uncertainties a series of experiments were conducted in which ice crystals collided with simulated hailstones over a wide range of controlled conditions. They are described in the following sections.

2. EXPERIMENTAL APPARATUS AND PROCEDURES

In the main experiments, a cloud of ice crystals was grown from vapour in the chamber illustrated in Fig. 1 and was drawn past stationary ice targets. The electrical current to the targets was measured as a function of stream velocity, temperature, temperature difference, applied field strength, surface structure of the targets and purity of the target ice. The experiments were conducted within a cold-room of operational temperature range 0° to −20°C.

Considerable effort was devoted to the development of a technique for the production of reproducible clouds of ice crystals, of concentrations sufficiently low to prohibit excessive aggregation rates of crystals upon the targets. In the technique eventually adopted water vapour from a steam generator was passed into the cold-room through pipes which were electrically heated and lagged to maintain their temperature above 100°C. The vapour could be directed into the cloud chamber or out of the cold-room again by the switching of solenoid valves. It could be introduced into the chamber in pulses so that the chamber did not become excessively heated and the quantity introduced could be strictly controlled. The chamber was divided by a door into two portions, both of which were lined with saturated blotting paper to help maintain high humidity. Vapour was introduced into the smaller, upper one whose volume was 0.05 m³, and allowed to cool to near ambient temperature, condensing as it did so into a cloud of small, supercooled water droplets. Efficient mixing and cooling were facilitated by fans. A metal rod, chilled by immersion in liquid nitrogen, was quickly plunged into and removed from the cloud chamber so that a small proportion of the droplets were nucleated. Ice crystals grew rapidly by vapour diffusion, the supersaturation with respect to ice being maintained by the residual water cloud. The droplets evaporated as the crystals grew. The longer the seeding time, the greater was the number of droplets nucleated and therefore higher concentrations and smaller crystals

![Figure 1. The experimental arrangement employed in the charge transfer studies.](image-url)
were produced. After a few seconds the door was opened and the two portions of the chamber were joined to form a single chamber of volume 0.35 m³. The cloud was thus considerably reduced in density as it mixed with the clear air of the lower chamber. It was left for a couple of minutes, during which time the liquid water became completely exhausted and the cloud acquired the temperature of the cold-room. The wholly-ice cloud was then drawn out of the chamber past the simulated hailstones. If it was not drawn out it remained in the chamber for about 20 minutes before evaporating and sedimenting out completely. The crystal concentration in the cloud, $N$, was governed largely by the quantity of vapour initially introduced and the residence time in the chamber. Values of $N$ ranging from 10 to 2500 cm$^{-3}$ were employed, but for most experiments $N$ was in the region of 300 to 1000 cm$^{-3}$. Because it was considered necessary rigidly to exclude liquid water from the cloud drawn past the targets it proved impossible reproducibly to generate large ice crystals, and the experiments were conducted with crystals of diameter around 10 μm. However, through judicious control of the seeding process and the rate of vapour input to the chamber it was possible to produce a small range of mean sizes, as illustrated in Fig. 2. Most of the ice crystals employed in these experiments were plates and columns, which are indicative of humidities rather below water saturation.

The cloud was drawn up through three 22 mm diameter brass tubes at velocities between 1.5 and 20 m s$^{-1}$. Two of these tubes contained ice-coated, metal targets and will be referred to as interaction tubes. The third contained a particle sampler which was a means of isolating a short length (80 mm) of tube from the airflow, so that the crystals in it were trapped and fell on to a formvar-coated glass slide. When the plastic had set, the replicas of the crystals were examined and their concentration and size distribution determined.

In a particularly detailed series of experiments, in which great care was taken to follow an identical procedure for cloud manufacture, it proved possible to produce and draw through the interaction tubes clouds of mean crystal diameter between 7.5 and 8.5 μm and concentration 1070 cm$^{-3}$ with a standard deviation of 320 cm$^{-3}$. These characteristics did not vary significantly with the air velocity. It should be remembered that only a very small volume of the cloud was sampled in each experiment. The crystal concentration was effectively averaged over about 10 ms while the currents were averaged over 10 s. For
calculations of the errors in the charge transferred per collision the relevant concentration variance is that for 10 s means, which would almost certainly be less than the one used here. The crystal concentrations varied considerably over a formvar slide. To ensure that a representative number of crystals were counted on each 19 mm diameter circular slide, two traverses with the microscope were made across each slide parallel to, and 3 mm either side of, a diameter. In general a third traverse was made perpendicular to the other two. In this way, some 14000 crystals were counted, in a total of 48, 18 mm long 0-27 mm wide strips on 17 slides obtained from 17 similar clouds to obtain the average crystal concentration. In the remainder of the experiments appreciable effort was devoted to the production of reasonably reproducible crystal clouds but greater fluctuations were considered acceptable. In general, in these experiments, conditions were reproducible over a day, or sometimes two or three days. When the apparatus was set up again, conditions were never quite the same. Comparisons between charging in different situations were only made when we were confident that the clouds in each of the experiments were similar. As it was necessary to leave the apparatus for about 30 minutes between experiments to enable the temperature of the target to return to ambient and the remnants of the previous cloud to evaporate, the number of experimental points which could be obtained from any series of experiments was limited.

There were two interaction tubes so that the charging of targets due to the collision of ice crystals could be compared for different conditions in the interaction region. It is likely that the ice crystals became charged during collisions with each other and with the walls of the tubes. Some of the charging of the targets would have resulted from direct transfer of these charges. It is very difficult to measure small charges on small crystals but their effect can be largely eliminated by considering differences in the charging of ice targets in different conditions.

The targets were 10 mm long brass rods of diameter 1-6 mm with a 0-2 mm thick coating of ice. They were positioned in the tubes so that the axes of the targets were perpendicular to the axes of the tubes. The rods were separated from the earthed tubes by PTFE insulators which were not exposed to the crystal flux. Cylindrical rather than spherical targets were used because a 2 mm diameter sphere would have presented a cross-sectional area to the crystal flux similar to any conceivable support for it, so that the charging of the support might have been of the same order as that of the ice. The targets were connected to earth through resistors in the range 0-1 to 10 GΩ and the potential difference across them was measured with a ‘Vibron’ electrometer whose input resistance was greater than 5 TΩ. The total capacitance to ground of probe, cable and electrometer was about 100 pF so that the response time of the electrometer to a step-function of current was between 10 ms and 1 s depending on the input resistor selected. The voltage measured by the electrometer averaged over a time long compared to this was therefore proportional to the current flowing into the ice target resulting from the many thousands of ice crystal collisions per second. The output of the electrometer was displayed on a pen or ultraviolet recorder.

The targets could be heated radiatively by a 150 W lamp containing a parabolic reflector placed close to a glass window in the interaction tube. Direct heating of the outside of the tube was avoided with an asbestos heat shield. Ice absorbs very little radiation at the frequencies for which glass is transparent so it appears that the brass absorbed heat and it was conducted to the ice surface. The temperature of the ice surface could be increased by up to 12 K above the airstream temperature. The probe temperature was measured with a very fine (40 s.w.g.) copper-constantan thermocouple which was attached to the middle of the underside of an ice target. The relationship between temperature elevation, ventilation speed and voltage applied to the lamp was determined in a separate experiment.
so that the thermocouple did not interfere with the charging. This was done prior to each set of charging experiments, a typical set of calibration points being displayed in Fig. 3. The thermocouple leads were shaded by the target from the direct heat of the lamp.

A shutter upstream of the targets enabled them to be ventilated by air from the cold-room to bring them into equilibrium with it before the cloud was drawn past them. Targets of various metals could be used instead of ice-coated ones.

It was necessary to take some care in the production of the target ice. The electrical properties of ice depend strongly on purity. The ice used was prepared from distilled, de-ionized water of conductivity $100 \mu\Omega^{-1}m^{-1}$ at $+20^\circ C$.

The brass targets were cleaned with fine-grain emery paper and wiped with a methylated spirit or 'Inhibisol' impregnated tissue to remove any grease or particles from the emery paper. They were then washed in pure water, wiped with a clean tissue and cooled to $-10^\circ$ or $-15^\circ C$ in the cold-room. They were then repeatedly dipped into freshly de-ionized distilled water at between $0^\circ$ and $2^\circ C$, so that a smooth layer of ice about 0.2 mm thick was formed on each. Fresh ice was made every day and it was always less than 12 hours old. In some experiments designed specifically to investigate the influence of the surface structure of the target ice upon charge transfer, rimed or frost-covered surfaces were produced.

The flow rate through the tubes was controlled by varying the voltage applied to the suction pumps and was measured — in each tube — by means of flow meters. Thermocouples were used to measure the temperature at various points in the apparatus to $\pm 0.3$ K.

A few experiments were performed in which the target ice, which was smooth, was doped with either HF or NH$_3$. This ice was made by dipping the brass targets into solutions of the impurities, varying in strength from $10^{-3}$ to $10^{-1}$M, at temperatures close to $0^\circ C$. The concentration of the impurities in the ice was much lower. It was determined by melting the ice targets, by dipping them into a known quantity of water, and measuring the concentration with ion-sensitive electrodes. It was not possible to dope the ice crystals.

In order to investigate the role that surface-potential differences might play in the
charge transfer a number of experiments were performed in which ice crystals were drawn past targets of various metals, which were not coated with ice. These experiments are described in section 4.

In the great majority of experiments the temperature differences between the interactants were created by radiative heating of the target, as already described. However, in some experiments the target was cooled by immersion in liquid nitrogen for about 10 minutes. Upon removal, it became coated with a layer of fine and delicate frost crystals. The temperature of such a target at the beginning of a charging experiment could be controlled by varying the interval between removal from the liquid nitrogen and exposure to the crystal stream. Alternate heating and cooling of the target ice relative to the crystals could be achieved by periodic irradiation of a chilled target which was connected to an effective heat-sink.

A number of checks were made to ensure that the measured charging effects were a consequence of collisions between ice crystals and the target, and not to some spurious phenomenon. Before every experiment air from the cold-room was drawn past the targets, both to bring them into thermal equilibrium with the airstream and to check that no current was detected by the electrometer in the absence of ice crystals. At the end of an experiment the crystal flow was usually stopped abruptly with the shutter so that cold-room air again flowed past the targets. The current decayed to zero with the instrumental relaxation time. No currents were detectable with the lamp on and crystals absent.

During a series of experiments ice crystals accumulated on the tube walls from which aggregates were sometimes blown. Some of these hit the targets, resulting in spurious traces. These were in the form of spikes indicating large charges being transferred in single events. They were easily distinguishable from the continuous currents produced by the impaction of, typically, $10^4$ to $10^5$ small crystals per second. Nevertheless, the whole apparatus was defrosted every two or three days to avoid these spurious effects. The first five seconds of each trace was ignored, because the charging often changed rapidly in a not very reproducible fashion, probably due to differences in the initial cloud characteristics. The charging therefore occurred only in the presence of ice crystals. Water droplets were excluded from the clouds. When all-water clouds were used no currents were detected. The charging was independent of the nature of the metal on which the target ice was formed.

The currents were estimated from measurements of the potential difference across a resistor between target and earth. In order to check that this voltage was not affecting the charging current, various input resistors between $50 \, \Omega$ and $10 \, \Omega$ were used to measure the current when similar clouds were drawn past the target in similar conditions. The current was found to be independent of voltage in the range used, which was $1$–$100 \, \text{mV}$. Taking the conductivity of ice at $-15^\circ\text{C}$ to be as low as $10^{-8} \, \text{mho m}^{-1}$, the resistance between ice surface and metal was only $300 \, \Omega$, so the proportionality between electrometer voltage and input resistance implies that the potential to which the ice was raised during the experiments did not affect the charging. However, a space-charge would be expected to build up in the ice because of impeded charge carrier exchange (proton-electron) at the ice–brass interface, and this might eventually reduce the charge transfer from the colliding ice crystals. The change in current following heating did not depend on how long after the start of the experiment the heating occurred (except when the crystal flux had declined) or on whether the current changed sign, remained positive but decreased, or remained negative and increased, as a result of the heating. This suggests that space-charge accumulation was not a problem. The charge transfer measured in these experiments would, therefore, have been the same as that to an initially uncharged hailstone under the same conditions.
In the absence of an airflow the heat from the lamp melted the ice when the thermocouple registered 0°C. A similar ice target without a thermocouple attached was melted by the lamp in the same time. When the lamp was run at full power and placed close to the target, it was just possible to melt the tip of the ice target in an airflow of 7 m s⁻¹. This occurred when the thermocouple registered −3°C. The thermocouple was attached to a part of the target which did not melt. Targets with and without thermocouples attached both had their tips melted in the same period of time. It was concluded that the thermocouple provided a reasonable estimate of surface temperature. It can readily be shown that the irradiation would have a negligible effect on the temperature of ice crystals in the air stream.

3. RESULTS OF THE ICE-ICE CHARGING EXPERIMENTS

Typical examples of measured currents to an ice target at the same temperature as the crystal stream to which it was exposed are presented in the curves A in Fig. 4. It is seen that after a few seconds a fairly constant positive current was achieved which persisted, with some fluctuations, throughout the duration of the run, which was generally around one minute: the fractional depletion of ice crystals within the chamber was small over this period. No significance can be attached to the sign and magnitude of this ‘control’ current since it is probable that contributions were made to it, in unknown proportions, both by charging resulting from ice–ice contact and by direct transfer to the target of charge acquired by crystals on colliding with the interior walls of the brass tube. The existence of this second process was confirmed, in subsidiary experiments, by employing a brass tube, the surface of which had been modified by heat treatment, and finding substantial changes in the control current. However, the influence on the charge transfer resulting from ice–ice contact of the temperature difference between the colliding specimens can validly be studied by comparing the control currents A in Fig. 4 with the current curves B, which relate to targets which had been heated by irradiation. For example, we see in Fig. 4(a) that the application of 10 V across the lamp was followed – after a few seconds required for the lamp to warm up – by a reversal of sign in the current and the achievement of a

![Figure 4](#).

Figure 4. Measured variations with time of the current flowing to (A) the control target and (B) the heatable target in two experiments: (a) in which the lamp was turned on and off once; (b) in which a series of voltages was applied to the lamp.
constant negative value. When the lamp was switched off the current slowly achieved roughly the same value as that in the control tube. Thus we see that warming of the target ice caused it to become more negatively charged. This pattern was repeated in Fig. 4(b), in which we see also that increasing the lamp voltage increases the current difference by an amount that decreases with increasing voltage increments. These observations were consistent in a series of experiments.

The temperature of the ice target was increased three times during each of a number of experiments. The change in the average charging current from that to an initially unheated target was measured each time, and corrected for any change in the current to the unheated target over the same period. The change in current was normalized by setting it equal to one for a temperature difference of 2.5 K so that the results of different runs could be compared. The results of six runs are combined in Fig. 5. The charging current rises to a plateau and is independent of temperature difference when this is greater than about 1.5 K. This was found to be true irrespective of the order in which the various voltages were applied to the lamp. Detailed studies of the effect on the current of heating the target ice
were performed at either $-10^\circ$ or $-15^\circ$C at velocities of 7.1 or 9.4 m s$^{-1}$. A similar relationship between charging and temperature difference was found in all of them. In experiments at higher velocities, up to 18 m s$^{-1}$, it was also found that once the target was heated sufficiently for a change in current to occur, further heating had no effect.

The average charge transfer per collision, $q$, resulting from the heating of the target ice, can be deduced from the measured difference, $I$, in the currents to the heated and control specimens by means of the equation $q = \frac{I}{(\alpha_1 N AV)}$, where $\alpha_1$ is the collision efficiency, $N$ is the ice crystal concentration in the airstream of velocity $V$, and $A$ is the cross-sectional area of the target exposed to the flow. If we take $\alpha_1 = 1$ (this assumption, together with a discussion of separation probability, $\alpha_2$, is developed in the following section) we find that a typical value for $q$ resulting from heating a target by several degrees, with $V = 6.5$ m s$^{-1}$, was about 20 aC for ice crystals of mean diameter around 8 $\mu$m. Some tentative evidence suggested that increasing the mean diameter by a factor of about 1.5 increased $q$ by a factor of about 5. The estimated error in these and subsequent values of $q$ was $\pm 20\%$.

A series of experiments was performed to investigate the effect of the impact velocity $V$ upon the charge transfer. In each experiment it was arranged that the heated target achieved a temperature at least 2.5 K above that of the control target, thus ensuring that the saturation value of $I$ was obtained. A set of results covering the velocity range 1.5 to 8 m s$^{-1}$ is presented in Fig. 6. It is seen that the charge transfer $q$ increases rapidly for values of $V$ above about 4 m s$^{-1}$. At lower velocities no significant current differences were measured. A further set of measurements was taken over the velocity range 5 to 18 m s$^{-1}$, and the strong dependence of $q$ on $V$ was confirmed, the results being displayed on curve 1 in Fig. 7. The charge transfer is seen to increase by about two orders of magnitude over this velocity range, the slope of the best straight line being $3.75 \pm 0.25$. This result was so striking that the entire set of experiments was repeated, the measurements being presented on curve 2 of Fig. 7. The slope of this line is $4.53 \pm 0.23$. Thus we conclude that for the conditions stated the charge transfer increases as roughly the fourth power of impact velocity.

**Figure 7.** The measured relationship obtained in two separate series of experiments (1 and 2) between $q$, the average charge per collision (arbitrary units, assuming $\alpha_1$ constant) and airstream velocity $V$. 
Figure 8. The measured variation with time of the current, $I$, flowing to a chilled frost target in two separate experiments, (a) and (b). The target was heated, by irradiation, in the time intervals A–B and C–D.

The rather complicated and sometimes inconclusive results of the experiments in which ice targets of a variety of surface structure were heated or cooled are described fully by Marshall (1976), and are here treated only cursorily. Fig. 8 shows that chilling a target to a temperature below that of the ice crystal stream caused it to become more positively charged. The increase in current appeared to be independent of the magnitude of the temperature difference, provided that this was in excess of about 1 K. It was found that whatever the nature of the crystal surface, heating caused it to become more negatively charged on exposure to the crystal stream, and cooling produced more positive charging. The most surprising feature of this set of experiments in view of earlier work was the insensitivity of the charging to the nature of the target surface. The charging of a smooth surface was of the same order as one made rough by riming, crystal aggregation or growth from the vapour.

The results of a series of experiments in which the charging of pure and doped target ice were compared are shown in Table 1. When the ice was doped with HF, the current to it was more negative than that to pure ice, and when doped with $\text{NH}_3$ it was more positive. In both cases stronger solutions increased the difference in currents. This behaviour is what one would expect from the theories of conduction in ice.

<table>
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<th>Description of target ice</th>
<th>Impurity concentration</th>
<th>Current to target (pA)</th>
<th>Current change, relative to control (pA)</th>
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<tr>
<td>Pure ice, heated</td>
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4. **Experiments on Discharge Limitation, Collection Efficiencies, and Ice–Metal Collisions**

Perhaps the most unexpected result of these charging experiments was that although the charge transfer was dependent upon the temperature difference between the colliding ice particles up to a value of about 1.5 K, it remained roughly constant thereafter. In principle, this saturation effect could have been caused by some blocking process in the ice itself or by a discharge between the interactants after separation. The latter appeared unlikely in view of experiments with HF-doped ice in some of which the negative charging was more than twice that of a heated pure ice target. Nevertheless, experiments were performed in an attempt to detect discharges between ice crystals and the target.

The first attempt was based on the ion-counter technique employed by Latham and Stow (1967) in experiments on snowstorm electrification. If micro-discharges were occurring between separating ice particles it would be expected that the ion concentration in the tubes would be increased. It was found that the current to the collecting electrode of the ion counter increased strongly if crystals were introduced into the airstream. However, the current was unchanged by the application of heat or the removal of the ice target and increased markedly with an increase in the voltage across the electrodes. These and other observations combine to demonstrate that the ion counter was probably measuring spurious currents resulting from collisions of ice crystals with the collecting electrode. This technique is not suitable for the measurement of ion concentrations in the presence of small particles.

The second method employed for the detection of discharges involved an RCA 9659R photomultiplier. This was placed as close as possible to the target ice in order to maximize the receipt of radiation. No light could be detected when ice crystals were drawn past an ice target which had been recently formed by immersion in water, heated with a lamp some 20 s earlier, formed from a $10^{-3} \text{M}$ solution of HF or formed by frostling after

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![Figure 9](image1.png)

**Figure 9.** Aggregates formed on initially smooth ice targets when exposed to identical numbers of ice crystals at four different values of $V$, the airstream velocity. A: $V = 1.9 \text{ ms}^{-1}$. B: $V = 3.1 \text{ ms}^{-1}$. C: $V = 5.3 \text{ ms}^{-1}$. D: $V = 18 \text{ ms}^{-1}$. 

removal from liquid nitrogen three minutes earlier. Experiments were conducted at velocities of 7 and 18 m s\(^{-1}\). It was also not possible to detect any light when large ice particles, formed by grating a block of ice, were poured into the interaction tube and drawn past the ice target. The output of the photomultiplier went off-scale in response to reflected light from a small torch or a spark from a conductor raised to a high potential.

The current from the photomultiplier in the darkened cold-room was 1.5 nA, which was close to the rated dark current for the tube. It was considered that changes in output current of 0.25 nA could be detected. This corresponded to 1.25 \(\times\) 10\(^4\) photons reaching the cathode per second. It was deduced from the geometry of the system that this was equivalent to about 30 photons per interaction. It was expected that at least one photon would be created for every electron which was transferred in the discharge. As the final charge per collision was about 120 electrons, the quantity of charge involved in the discharge would have been greater than this, or of the same order, if the original charge transfer had been proportional to temperature difference.

These experiments indicate that the plateau observed in the current/temperature curves was not a result of discharge limitation.

A surprising result of the charging experiments was the dramatic increase in charge transfer with airstream velocity: \(q\) was found to increase 200-fold when \(V\) increased from 5 to 18 m s\(^{-1}\). Marshall (1976) presented arguments showing that this sensitivity of \(q\) to \(V\) is unlikely to be primarily a consequence of variations with velocity of contact area or collision efficiency, \(\alpha_1\). He performed some experiments, which are now described briefly, to test the hypothesis that variations in the separation probability, \(\alpha_2\), could have been responsible.

Fig. 9 presents photographs of crystal aggregates on initially smooth ice targets which had been exposed to similar crystal clouds at various velocities. The product of exposure time and airstream velocity was constant for all situations, so true comparisons can be made between the aggregates produced at all values of \(V\). The measured dimensions of
these aggregates, and ones obtained in other, similar, experiments, are presented in Fig. 10. They are seen to decrease substantially as the velocity increases. In further experiments, employing a target with four times the surface area, the total mass of the ice crystals collected was measured for a range of velocities. The results showed that the collection efficiency, \( \alpha_1(1 - \alpha_2) \), at 18 m s\(^{-1}\) was roughly 1/10 of that at 5 m s\(^{-1}\), the estimated value at this lower velocity being between 0.6 and 1.

These crude experiments suggest that variations in separation efficiency probably played a significant role in the observed \( q - V \) dependence. However, it is unlikely to be the only important factor. The charging experiments of Buser (1976) and also of Latham and Miller (1965) indicate that the charge transfer per rebounding event, \( q' (= q/\alpha_2) \), can vary substantially with velocity.

In an effort to establish the role of surface-potential differences on the charge transfer resulting from ice-ice contact, a series of experiments was performed to investigate the charging which occurred when clouds of ice crystals were drawn past targets of various metals, which were not coated with ice. The differences in the surface potentials of the metals were measured before and after each experiment by the Kelvin ionization method. The metal target was placed centrally in a slightly wider brass tube and the air gap between them was ionized with \( \alpha \)-particles from an americium source. Because of the difference in the surface potential of the two metals a current flowed between them through the ionized atmosphere. This current could be stopped by applying a back-off voltage equal to the difference in surface potential between the target and tube. Measurement of this voltage thus gave the difference in surface potential between the target metal and brass.

Probes of brass, aluminium, nickel, platinum and magnesium were used. Because of the humid environment inside the cold-room the metal surface almost certainly absorbed water vapour. The difference in surface potential declined within a few minutes of being placed in the cold-room, and by the time they had reached thermal equilibrium there was no significant difference between surface potentials of brass, platinum and nickel, and only a small, steady one (~100 mV) between these and aluminium and between aluminium and magnesium.

When the interaction tubes contained targets of different metals and crystals from the same cloud were drawn past each, the metal with the more negative surface potential (or higher work function) was charged more negatively. Usually there was a positive current to magnesium, a negative one to brass and a smaller current of either sign to aluminium. Nickel and platinum behaved similarly to brass. Sometimes the currents to both magnesium and any one of nickel, platinum or brass were of the same sign, but the first charged more positively than any of the others in all 36 half-minute experimental runs. Typical examples of the current traces (which have been smoothed by plotting two-second means) are shown in Fig. 11. These refer to clouds of crystal concentration 500 cm\(^{-3}\) being drawn at 9-5 m s\(^{-1}\) past 1 mm diameter metal probes. The average charge \( q' = q/\alpha_2 \), transferred in a rebounding collision per volt difference in surface potential is about 2 fC V\(^{-1}\) if \( \alpha_1 = 1 \)

![Figure 11. The measured variations with time of the current, \( I \), flowing to targets of different metals in two experiments (a) and (b). A, magnesium; B, brass; C, aluminium.](image)

Figure 11. The measured variations with time of the current, \( I \), flowing to targets of different metals in two experiments (a) and (b). A, magnesium; B, brass; C, aluminium.
and we take $\alpha_2 = 0.9$. This is reasonably consistent with the value of $15 \text{fC} \text{V}^{-1}$, obtained for collisions at $-45^\circ \text{C}$ by Auf der Maur and Buser (1974), when account is taken of the fact that the diameter of their ice spheres was about three times that of the ice crystals used in the present work. It is noteworthy that the measured differences in the currents to different metals are of the same order as those to different ice surfaces.

Despite the correlation between charging and surface-potential difference, we are unable to corroborate Auf der Maur and Buser's conclusion that the charge carriers are electrons. It is very likely that the surfaces of our metals absorbed water and thus the charge carriers may have been protons and hydroxyl ions. Because of the problem of water absorption, experiments on ice-metal interactions were not pursued further.

5. Discussion

We now consider the extent to which the charge-transfer measurements described in this paper are explicable in terms of the surface-potential difference mechanism, proposed by Auf der Maur and Buser.

An ice surface formed by sublimation is believed to have a higher work function than one formed by deposition from the vapour, and will thus tend to be charged more negatively. In the saturated environment of an ice cloud, a warmer ice surface will start to evaporate, and deposition will occur on a colder one. Thus the warmer surface will acquire negative charge and a colder one a positive charge. The charge transfer will increase with increasing temperature difference until a plateau is reached. These qualitative predictions are consistent with the observations made in our experiments. The achievement of the charging plateau would not be expected to be sharp because the heating of the target was not uniform.

The similarity between the charging during ice–ice and ice–metal interactions suggests that the same mechanism was operating. The experiments described in the present work were hindered by water absorption on the metal surfaces, but different metals charged differently and the differences between the charging of these metals were similar to those between the charging of heated and unheated ice. Auf der Maur and Buser, working in a drier environment at $-45^\circ \text{C}$, did not appear to have this difficulty. They found a strong correlation between charging and work function for ice–metal interactions. The difference in charging between a heated and a cooled ice surface corresponded to a difference in work functions of 0.2 to 0.3 eV in both experiments, which is similar to the differences between the work functions of ice surfaces formed by deposition and evaporation, measured by Takahashi (1973). The experiments in which pure ice crystals collided with doped ice targets described in this study were explicable in the same terms as those performed by Auf der Maur and Buser, in which doped ice spheres collided with a platinum target.

If we assume that the collision efficiency $\alpha_1 = 1$, the mean charge transfer per collision when $V = 18 \text{ m s}^{-1}$ was $\approx 800 \text{ aC}$. Taking the separation probability to be about 0.9 (section 4) we see that the mean charge transfer per rebounding event, $q'$, was $\approx 900 \text{ aC}$. If we assume that the surface-potential difference between the ice crystals and the heated ice target was $100 \text{ mV}$ the theoretical value of $q'$ agrees with that measured if the area of contact on collision was $1 \mu\text{m}^2$, which appears reasonable.

It is perhaps worth noting that although the classical thermoelectric effect is qualitatively consistent with many of the observations made in these experiments it is quantitatively inadequate by five orders of magnitude. However we cannot discount the possibility of a more powerful thermoelectric effect associated with the surface layer of ice.

It is difficult to make a strict comparison between the charge-transfer measurements obtained in these studies with the wide range of values determined by other workers. In these latter studies the values of velocity, temperature, crystal size and other parameters
Figure 12. The variations of $q'$, the charge transfer per rebounding event, with ice crystal size, $d$, derived from the experiments of several workers. 1 Latham and Miller (1965); 2 Latham and Montagne (1970); 3 Latham and Stow (1967); 4 Reynolds, Brook and Gourley (1957); 5 Auf der Maur and Johnson (1972); 6 and 8 Church (1966); 7 Auf der Maur and Buser (1974); 10 Latham and Mason (1961a); 9 the present experiments. In 3, only a range of sizes was quoted. Marshall (1976) has argued that point 8 is probably underestimated by about one order of magnitude.

Varied substantially, so quantitative agreement would not be expected. However, it is instructive to examine the relationship between crystal size and charge transfer per collision, derived from the various experiments, presented in Fig. 12. With one exception, it is seen that over a range of crystal diameter, $d$, from about 10 to 300 $\mu$m the relationship between $q'$ and $d$ is described well, considering the experimental differences, by a square-law. This is consistent with the predictions of the surface-potential difference mechanism. The very low value of $q'$ measured by Latham and Mason (1961a) is so inconsistent with the collection of other values displayed in Fig. 12 that it should probably be discarded. The levelling-off in the curve for values of $d$ in excess of about 300 $\mu$m is to be expected, since the contact for irregularly shaped ice crystals of this size colliding with hailstones could not be so intimate as that for smaller crystals. Thus it appears that the charge transfer per rebounding collision, for ice crystals of sizes covering the range likely to be of importance in thundercloud electrification, may be expressed roughly by the equation

$$q' = 5d^2$$

where $q'$ is measured in attocoulombs and $d$ in micrometres.

The field growth calculations of Illingworth and Latham (1977) indicate that under a non-inductive ice–ice charging process governed by Eq. (1) field growth will occur sufficiently rapidly to produce breakdown fields in thunderclouds within the available time.

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