The effects of surface curvature and temperature on charge transfer during ice-ice collisions.

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Key Points

Charge transfer between ice surfaces, of differing curvatures, in under-saturated and near-saturation environments.

Conditions for the existence of a quasi-liquid layer disappear at point of contact; a new layer must be established about bridging spicules.

Inference is that mass transfer in a quasi liquid layer is parallel, not transverse, to the interface, driven by differences in curvature.

Abstract

Charging of colliding ice matched somewhat a study (Mason & Dash, 2000, https://doi.org/10.1029/2000JD900104) in which there was qualitative agreement with a prediction of Baker and Dash, (1989, https://doi.org/10.1016/0022-0248(89)90581-2 1994, https://doi.org/10.1029/93JD01633) based on the proposal by Turner and Stow (1984, https://doi.org/10.1080/01418618408236549). The magnitude (a few picoCoulombs) of the transferred charge Q observed by Mason and Dash (2000) between two similar surfaces 0.5mm thick but of differing temperatures, raised doubt that charge transfer is solely a surface-layer effect in such non-equilibrium conditions. Here, during repeated collisions between an ice bead and a sheet each of order 3 mm thickness, the lack of dependence of charge transfer on common temperature, and the polarity of the pieces upon separation, is consistent with the earlier results over a greater range of temperature (235K to 268K) if the conjecture by Dash et al (2006, https://doi.org/10.1103/RevModPhys.78.695) that the mass transfer could be driven by differences in curvature. The positive charge would be transported in the mass carried along the QLL on the surface of connecting spicules. The transfer of kinetic energy is estimated to be a factor of 10^4 greater than in the study by (Mason & Dash, 2000, https://doi.org/10.1029/2000JD900104) such that this result constrains the effect of postulating (Dash et al, 2001, https://doi.org/10.1029/2001JD900109) collisional melting.

Index Terms: 3320 Cloud Physics 3304 Atmospheric electricity

Key Words: ice surfaces, charge carriers, charge transport, growth processes, interfaces, quasi liquid layer, topological diode.

1. **Introduction**

As a possible mechanism for charging of thunderclouds, the charging of ice particles has been studied in increasing detail for nearly seven decades (e.g. Reynolds et al., 1957; Takahashi, 1978; Gaskell & Illingworth, 1980; Jayaratne et al., 1983; Turner & Stow 1987; Baker et al., 1987; Keith & Saunders, 1989; Saunders et al., 2006), but the physics of the charging process has still not been explained. The charge separation Q appears to depend on the growth states rather than the temperature difference between colliding ice surfaces (Buser & Aufdermaur, 1977; Gaskell & Illingworth, 1980; Baker et al., 1987; Jayaratne, 1993). Baker et al. (1987) concluded that during diffusion the faster-growing ice surface becomes positively charged. This empirical observation has since been supported by the results of Luque et al (2016) and Emersic & Saunders (2020) although**,** as the latter note, the "qualitative empiricism … is not capable of providing quantitative predictions." As in much earlier work, these studies attempted to model the cloud-conditions of small (order of 10 micron) crystals colliding with a replica of graupel that is larger (mm size) ice and with supersaturation and the presence of liquid water as droplets).

Turner and Stow (1984) suggested that Q might be related to the quasi liquid layer on ice (Fletcher, 1968; Turner 1982; Kling et al., 2018). Baker and Dash (1989, 1994) (hereafter referred to as BD) proposed that the charge is transferred through a liquid-like surface layer. In the BD model, mass is transferred from the thicker layer to the thinner and in effect a nett charge is carried within the transferred material. Mason and Dash (2000) (hereafter, MD) designed a test of the basic prediction of the BD theory with the control of mean temperature and growth rate while applying a temperature-difference between two otherwise identical flat ice surfaces undergoing repeated collisions.

MD tested whether mass exchange accompanies charge transfer by studying the successive impacts between two ice-covered quartz crystals, one driven by an acoustic loud-speaker. The controlled variables were the environmental temperature, temperature difference between ice-plates, rates of growth and evaporation, and the time between events. Ice surfaces were formed by vapour deposition on the quartz to a depth of about 0.1mm and up to 2mm in lateral directions, and were separated by about 0.3mm. The results from recording individual collisions showed that mass transfer was approximately proportional to, and accompanied, charge transfer. The faster growing ice plate became positively charged in the range studied, 253 K to 273 K. The estimated irregular roughness of each surface was about 0.03 mm.

While the correlation between mass and Q followed the prediction of BD, the sign of Q, the separated charge between the surfaces did not match their prediction, and the size (a few picoCoulombs) of the transfer was several orders of magnitudes greater (tens of femtoCoulombs) than that reported by BD in their earlier work.

MD concluded that the highly non-equilibrium condition of the temperature-difference driving the mass transfer across the interface during a collision may be the reason for the disagreement with earlier results and with the BD theory, which is based on a QLL thickness under equilibrium conditions and on the vapour-diffusion driven difference in the QLL of each surface approaching collision.

Here, we report results of an experiment in which two different surfaces collide repeatedly at common temperatures and, with respect to the flat surface, in an under-

saturated and near-saturated atmosphere. While the original motivation of all such work concerned the understanding of the natural phenomenon of lightning, the fundamental physical properties of ice, including mechanisms of charge transfer, can be elucidated only by minimizing or controlling the number of confounding variables. As in MD, this work is without the presence of liquid water for as Emersic & Saunders (2020) noted riming adds additional complexity, but contrary to MD without the temperature-difference that could drive the mass-transfer across the interface at collision. Thus contrary to MD, and also to particle experiments such as Emersic & Saunders (2020), the ice-surfaces are macroscopically in thermal steady-state. Similarly to MD, the collision-speed (at about 1 ms^{-1}) is up to an order of magnitude slower than the earlier studies.

1. **Experimental design and procedure**

As shown in Figure 1, the apparatus consisted of a solenoid connected to a lever which activated a pendulum. The pendulum consisted of three components: a swinging aluminium bar, at one end of which was screwed a brass plug into which a nichrome wire (diameter 1 mm) was soldered. The whole assembly (90 mm in length) was mounted on an aluminium post. An aluminium plate (30 mm x 20 mm x 5 mm) was mounted onto a TEFLON™ post to one side of the aluminium post. This plate, when iced, served as the end-stop for a bead of ice on the tip of the nichrome wire of the pendulum.

The aluminium post and the TEFLON™ post were attached to a circular plate which formed the base of a cylindrical container of inner diameter 210 mm and height 280 mm. O-ring seals were located in the bottom plate and in the top flange of the cylindrical shell so that the whole stainless steel assembly could be evacuated.

Inside the cylinder, in a metal box, the charge meter interface (which employed a CA3130) had an effective integrating capacitance of 1 pF across a 1 TOhm glass-walled resistor, giving an overall time constant of 1 s. In operation, the A.C. noise was 5 mV, equivalent to a minimum charge resolution of 5 fC. A solid-state pressure sensor (accuracy ± 2 mbar) was installed within the cylinder, together with a sensor suspended from the top plate to measure air temperature (accuracy ± 0.5 K; precision to 0.1 K) in the middle of the cylinder.

The cylindrical assembly was placed inside a large cold room in which the temperature could be stabilized to within ± 1 K over a period of several minutes, thus minimising temperature gradients inside the cylinder. Connections to the cylinder were made through ports in the cold room, leaving the interior of the cold room undisturbed. A pulse generator connected to the solenoid enabled collisions at an interval of 3 s. This period was sufficient to allow the pendulum to come to rest between successive collisions.

Using the TEFLON™ post as a handle, the rectangular aluminium plate was repeatedly immersed in a beaker of cold water of resistivity 70 kOhm-m until the layer of ice formed was about 3 mm thick on the larger faces. Using the same water, the tip of the nichrome wire similarly acquired a tear-drop-shaped bead of ice approximately 3 mm in diameter. A small loop was formed at the end of the wire beforehand so that ice completely surrounded the end of the wire. The curvature of the ice bead was thus much greater than that of the plate.

Since the pendulum was connected to the case of the apparatus and the target was held virtually at earth potential via the chargemeter, both the iced plate and bead of ice were at a common potential. The situation may be compared to the method of Glass (1969) in that the working material must be electrically short-circuited and free to expand to remove the complicating presence of electric field and stress-induced polarizations, respectively. For both pieces of ice near earth potential, the advantages were that the state of both pieces of ice was well defined and it was unnecessary to attempt to measure the charge on the bead. With the plate free of ice, and connected to a voltage supply in series with a resistor, the contact time between it and the tip of the nichrome wire was determined to be 15 ms.

The experimental cycle was that, beginning at 268 K, the temperature was reduced in 10 K intervals to 228 K. The refrigerator thermostat could be set so that the refrigeration system did not operate during the course of any measurement. Thus, on repeatedly spraying the inner walls of the cylindrical shell of the apparatus until icing was complete, the region near the target surface became nearly saturated with respect to ice.

The speed of the bead of ice at contact is of the order of 1 ms^{-1}, as measured from video of the motion, and of the rebound 0.6 ms^{-1}, so the corresponding transfer of kinetic energy is 0.6 mJ (with an uncertainty of about 10%).

1. **Results**

The charge Q on the target is plotted as a function of temperature T for both an under-saturated and a saturated atmosphere (in Figure 2). The mean and standard deviation of Q was calculated for measurements over 6-minute intervals (about 120 contacts). The standard deviation in the data is greater than any apparent trend of Q with T in both states.

The negative sign of the charge on the flat target is the same in both saturated and under-saturated conditions at all temperatures. From the results of Buser and Aufdermauer (1977), Gaskell and Illingworth (1980), and Jayaratne *et al.*

(1983) the sign of the charge transfer was expected to reverse at some temperatures. In the MD results, the sign of Q on the similarly flat, stationery target was positive.

1. **Discussion**

The large magnitude (pC) of Q on the target in the ice-ice collisions reported here is a major difference between this work and most other work reviewed earlier. However, the magnitude does agree with the results of MD, and an apparent difference in the polarity of the transferred charge, in that MD's stationary plate charged positively, is explicable in terms of the quasi liquid film QLL on ice, as discussed below.

Since Turner and Stow (1984) proposed that the charge transfer is mediated by the QLL on ice, and models of that film show that the thickness decreases with decreasing temperature, then a lesser magnitude of Q with decreasing temperature could be expected because of a thinner QLL. However, in the present experiment, an apparent difference is in only the standard deviation of results from under-saturated to near-saturated conditions, which could indicate that even without temperature-dependence of Q, the surface condition is the controlling parameter, although the results converge near 270K.

In the extension by BD of the theory, during contact mass will move towards the surface of thinner QLL. Furthermore, the surface acquiring mass will become positively charged. Thus, the surface of initially thinner QLL will acquire a positive charge. Here, the flat plate charged negative. The saturated vapour pressure of a flat plate of ice is lower than for a convex ice bead, so in under-saturated conditions a flat surface will be decaying at a lesser rate than a convex surface (the Kelvin effect in solids). On energetic grounds, during evaporation the surface with greater sublimation will have the thinner QLL. Thus, during contact, the thicker QLL of the flat plate will lose mass to the convex surface, rendering the flat surface negative, as was observed.

Lowering the temperature of the apparatus thins the QLL on both surfaces, but the Kelvin effect is maintained so that the polarity of the charge transfer is unchanged. In the iced system, the atmosphere is nearly saturated with respect to the plate but the bead remains relatively the more under-saturated so the plate would still be expected to charge negatively. The magnitude of the Kelvin effect is not of issue, only that there is an effect that drives the process in one direction. This explanation for the polarity of charging is consistent with the conjecture by Dash et al. (2006) that the mass transfer can be driven by differences in curvature, and is thus without the need for postulating (Dash et al, 2001) collisional melting. The lack here of an imposed temperature difference (used by MD to stand in for a difference in vapour-diffusion so as to create a difference in relative thickness of the QLL), clarifies the process of charge transfer between colliding ice particles.

Because the QLL is thinner at lower temperature it is reasonable to infer that there will be a correspondingly greater variability if the concentration of available charge-carriers is reduced towards some threshold quantity, leading to the trend in variability of charge transfer observed here. This trend, together with observations of the magnitude of the charge transferred and its apparent dependence on ice saturation, supports the theory and observed magnitude of charge transfer found by Mason and Dash (2000), despite the considerable difference in the experimental methods used.

The contact time was 0.25 ms for MD and 15 ms here; a difference of a factor of 60. The charging rate, contrarily, was 8x 10^{-9} Cs^{-1} for MD yet only 2x 10^{-10} Cs^{-1} here; a difference of a factor of 40. The difference in gross contact area (mm^2 for MD, 10^{-2} mm^2 here) could compensate for the difference in order of magnitude of contact-time. However, a consideration of the structure of the contacting surfaces suggests that may be merely co-incidental. The consideration of the surface structure may also indicate why the results of many studies have a great variability.

In the experiment of MD, the estimated 50% transfer of kinetic energy was 10^{-8} J from which Dash et al (2001) noted a thickening of the QLL to 160 nm can be inferred if the collisional melting across the estimated contact area produces an increase of surface temperature of about 12 degrees. Consequently, Dash et al (2001) inferred that ionic and protonic diffusions would be comparable to that of liquid water.

In all such analyses, the principal assumption is that transfer is transverse to, that is across, the QLL of the colliding surfaces. Collisional melting would act to enhance Q on the basis that the QLL thickens, and that the thickness is proportional to energy transfer. Any model must explain both the empirical observation that the surface growing faster by vapour deposition charges positively, and the mass-transfer in MD being driven by temperature-difference.

Simulations (Furukada and Nada, 1997) of the creation of a surface by molecular dynamics show that the disordered surface layer (QLL here in ice) exists as a necessary transition layer between the vapour phase and the solid structure with a regular lattice (for convenience hereafter called bulk ice). In the absence of liquid water (that is, without any riming) when two pieces of ice contact one another, the previously existing two QLLs are sandwiched between bulk ice and thus there is no longer a vapour-solid interface, and the previously disordered QLL layers must re-orient.

Since the surfaces are not molecularly flat, necks of bulk ice must necessarily be formed. That is, spicules of height at least equal to the thickness of two QLLs. Not only will the spicules be of indeterminate diameter (although of at least several molecules) but also of indeterminate number, and both factors will vary with each contact within the macroscopic area of contact. During the contact, while the spicules exist, the necessary vapour-solid transition layer (the QLL) is a collar about the spicule (and of course continuous with the QLL on the exposed surface of each of the adjacent solid ice samples).

In that situation, as Dash et al (2006) proposed, a "[t]hermomolecular pressure gradient will occur in a liquid regardless of its construction" from which can be inferred that if the material in the QLL is sufficiently mobile, then with such a pressure gradient parallel to the interface between the QLL and the bulk ice, any mass transport will occur in the QLL parallel to that interface. That is, the flow is not across the QLL. Hitherto all models have been of the transport across the QLL, on the basis that in collisions there would be created a joint QLL that is sandwiched between the two bulk ices. Consequently, in this new model of flow in the QLL, the electrical effects in the surface are a result of the superficial molecular geometry and unrelated to the properties of the bulk ice, and in effect the QLLs in contact acts as a topological diode.

In the work of MD the temperature difference, which would create the thermomolecular pressure, was imposed externally upon the ice surfaces. MD estimated 1% coverage of the flat surface (that is, quoted 10^{-2} mm^2 in plates 1mm on a side) by raised parts about 0.03 mm in height. Assuming that the raised parts are approximately cylindrical, then there would be about 13 connecting spicules with a circumferential total of about 1.3 mm. For the rate of charge separation of about 8x 10^{-9} Cs^{-1} as reported in MD, and using the estimated surface charge density of 3 x 10^{-4} Cm^{-2} of Baker and Nelson (2002) then the speed of the flow across the circumferences (in total) has to be about 20 mms^{-1} which is in agreement with the superficial diffusion rate of 40 mms^{-1} that follows from the equalisation of charge distribution through a 40 nm film in the order of a microsecond estimated as necessary by Dash et al (2001).

The results here agree with those of MD. That is, Q is not temperature dependent, whereas models and measurements of the QLL (for example, Beaglehole and Nason, 1980) show the thickness of the QLL to be a function of T (approximately exponential from 10 K below 273). As Dash et al (2001) noted, the effect of the existence of the QLL would be to allow changes in surface charge distribution to occur with diffusion of ions at values near that of liquid water, and thus on time scales of about a microsecond (which is significantly less than the duration of collisions). In addition, they noted that Q transfer would occur at temperatures lower than those at which the QLL has been seen to exist, since collisions could contribute additional energy to disordering the surface (i.e. collisional melting).

Neither of these two effects introduce nor require a dependence of Q on the thickness of the QLL; merely the existence of the QLL greater than some minimum thickness. That the calculated energy transfer here is 10,000 times that of MD (and through a contact area about 10^{-2} of the size) yet the rate of charge transfer is 1/40 that of MD, adds to the supposition that the increasing depth of the QLL (that would follow from a theory based on collisional melting) is not a factor in Q transfer, at least not beyond some minimum value provided that the concentration of available charge carriers is sufficient.

A corollary of the work described here is that ice electrification within clouds need not invoke temperature effects alone; the charge transferred during ice-ice collision may also be a function of the difference in size (curvature) of the particles undergoing gravitational separation. These effects of course may be completely obscured in conditions where there are several confounding factors, such as liquid water and dissolved ions (in the liquid water or concentrated in the QLL), which diversity is consistent with the great variation in results reported from the natural environment.

All results combined indicate that topology of the ice rather than symmetries of the bulk material is important. Regardless of the method of creation of the surfaces, the essential results would appear to be stable regardless of any crystalline defects and so the superficial excitations must be stable even with such defects being present.

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Captions

Captions

Figure 1. Front elevation of the apparatus which was housed in a stainless-steel cylinder. A solenoid connected to a lever activated a pendulum. To scale: the inner diameter of the base-plate was 210mm.

Legend: 1. Base plate. 2. aluminium post. 3. aluminium slab. 4. container for charge-meter. 5. closed lid as a shield against electrical noise. 6. Aluminium post (main stand). 7. Solenoid. 8. Arm that triggers pendulum. 9. Sliding rod triggered by solenoid. 10. physical pendulum (3 parts: bar, screwed rod, nichrome wire). 11. a stop to limit swing of pendulum

Figure 2. The charge Q on the flat ice target plotted as a function of temperature T at various pressures for an under-saturated atmosphere, and for a near-saturated atmosphere.