Charge Separation Associated with Secondary Ice Crystal Production

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(Manuscript received 12 March 1979, in final form 15 June 1979)

ABSTRACT

Laboratory studies of rime growth on a moving rod under conditions of secondary ice crystal production show that the rod acquires a positive charge, equivalent to charge associated with each ejected particle of 5×10^{-4}C. Ice crystals produced by seeding also impart a positive charge to the rime, equivalent to a charge per particle of 5×10^{-16}C. As the water vapor supply is cut off, the charge sign reverses. The results suggest that the sign of the charge transfer depends on the physical state of the rime surface and its vapor pressure excess or deficit relative to the environment. Charge separation in convective clouds is critically dependent on the changing proportion of graupel and small secondary ice crystals.

1. Introduction

There is extensive field evidence that separation of electric charge in the atmosphere is associated with the presence of graupel (Kuettner, 1950; Latham and Stow, 1969). In order to elucidate the detail of the mechanism of charge separation, studies have been carried out to simulate graupel growth in the laboratory (Reynolds *et al.*, 1957; Church, 1966; Takahashi, 1978). The experiments showed that the presence of ice crystals produced by seeding colliding with the graupel was necessary before significant charge appeared, the magnitude of which was related to the cloud water content and the temperature excess of the graupel caused either by latent heat release of accreting drops, or by radiation. In the absence of ice crystals, no charge separation took place.

Recent laboratory studies have shown that under specific conditions, secondary ice particles are produced during graupel growth (Hallett and Mossop, 1974a,b; Mossop and Hallett, 1975; Mossop, 1978a,b). Ice crystals result from the buildup of rime on a rod moved by a rotating arm through a supercooled water cloud. The source of the crystals is thought to be the ejecta produced when the supercooled drops freeze on impact with the rod (Choularton *et al.*, 1978; Mossop, 1979). This occurs only within a narrow range of temperature (−3 to −8°C) at impact velocities >0.7 m s^{-1}, and only when cloud droplets of diameter >25 μm in concentration >0.1 to 1.0 cm^{-3} are present together with a suitable proportion (100:1) of drops of diameter <13 μm.

The experiments reported here were undertaken to find out whether secondary ice crystal production could be important in the charge separation process. This could occur directly, through charging of secondary particles as they are produced, or indirectly through collision of secondary ice particles with graupel particles already present or formed from the freezing of supercooled drops.

2. Experimental studies

The technique used in this study was similar to that used by Hallett and Mossop (1974a,b). An inner chamber of sheet aluminum 1 m high × 1.5 m × 1.5 m was constructed inside a cold room. A water cloud was produced by steam injected near the edge from a boiler below. Two vertical riming rods, 30 cm long, could be rotated about a vertical axis at speeds up to 3.5 m s^{-1} by a motor external to the cold room (Fig. 1). Higher speeds were unrealistic in this experiment as particles began to break off under the increasing centrifugal acceleration (>10 g). Cloud liquid water content was estimated from accretion rate and collection on an impactor slide; drop and crystal sizes were measured directly from particles collected and replicated in formvar.[1] Each riming rod consisted of a 3 mm

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[1] Crystals were collected by sedimentation; drops by moving a slide through the cloud from outside the chamber on a whip collector.

0022-4928/79/112230-06$05.50

diameter stainless steel tube mounted in paraffin wax at either end. One rod led to a charge amplifier ("gain", 1 mV = 10^{-13} A) mounted on the rotating shaft. Charge flowed to ground with a time constant of 1.0 s. Slip rings mounted externally to the cold room carried signals and amplifier power. Cups 5 cm in diameter were mounted at top and bottom of the riming rod to rime preferentially during rotation to prevent a rime bridge giving a path to ground (Fig. 2). Temperatures were measured by thermocouples in the cloud, with an uncertainty of ±0.5°C.

Ice crystals appeared in the cloud depending on whether the conditions for secondary ice production were satisfied. On first introduction of the water cloud, visibility was low (<1 m) with a large number of small drops formed on the local aerosol, indicated by a Pollak counter to be $\sim 10^4$ ml^{-1}. After about 1 h, the water being continually supplied into the closed inner chamber, the visibility improved considerably as the spectrum shifted to larger drop sizes. Operating conditions were selected for optimum secondary ice production. In practice this required conducting the experiments during the appropriate stage of evolution of the drop spectra, so that sufficient small (<13 μm) and large (>25 μm) diameter drops were present in the cloud. Background ice crystal concentration was $\sim 1\,\ell^{-1}$; secondary concentration reached a maximum of $\sim 100\,\ell^{-1}$. Crystals were viewed by looking nearly directly into the collimated beam of a microscope illuminator.

Charging measured in the presence of secondary ice particles increased with velocity $\geqslant 0.7$ m s^{-1} (Fig. 3, squares). Below this speed no crystals appeared above background and no charging could be measured. In all cases, the charge on the riming rod was positive. When the riming rod was stopped and restarted, the charge transfer fell rapidly (within a few seconds) to zero and then rapidly recovered to its original value (Fig. 4). This behavior occurred irrespective of the length of the rest period.

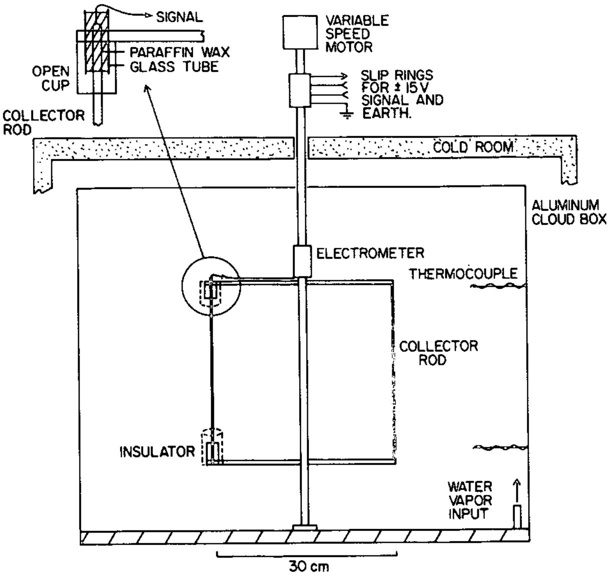

Fig. 1. Apparatus for rime growth and charge measurement showing detail of the riming rod insulation.

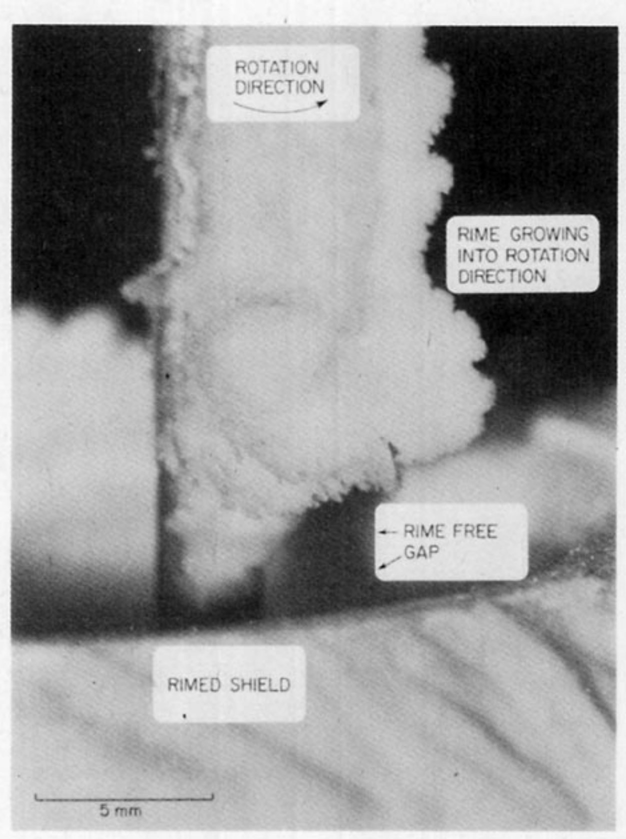

Fig. 2. Detail of rime growth on rod and one of the shields which prevented rime bridging and a short to ground.

There are two possible explanations for the origin of the charge transfer: 1) the ejected fragment removes the charge or 2) the riming rod sweeps out the ejected fragments and the charge transfer occurs during the subsequent collision and separation process. In order to help discriminate between these two possibilities, a further study was made of the charge transfer occurring when the riming rod moved through a cloud of ice crystals, similar to experiments carried out by earlier investigators (Reynolds *et al.*, 1957; Hobbs and Burrows, 1966; Takahashi, 1978).

Ice crystals were nucleated in the cloud by a liquid nitrogen cooled brass plate of area 20 cm^2, a copper wire of area 0.5 cm^2, or by popping a 5 mm plastic bubble of packing material in a syringe. Solid carbon dioxide seeding was avoided because of possible effects of dissolved gas on drop freezing during the riming and charge transfer process (Dye and Hobbs, 1966). With the arm rotating, ice crystals were dis-

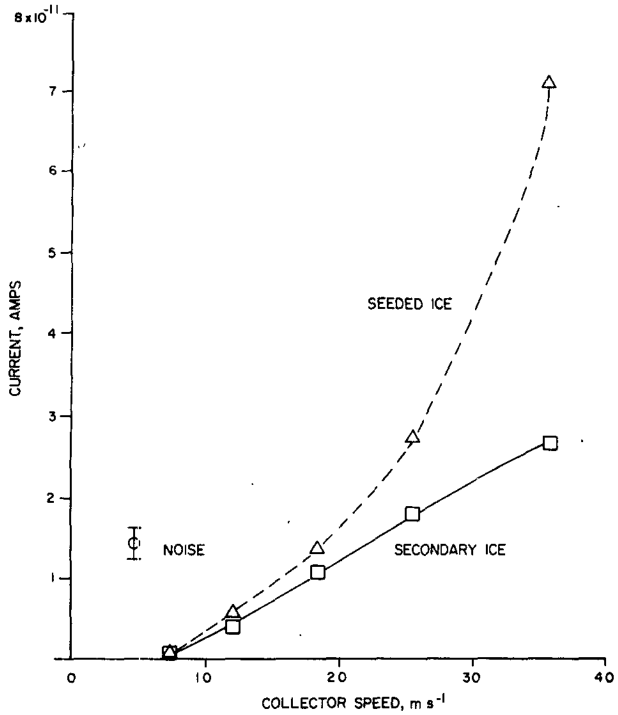

FIG. 3. Velocity dependence of charging with secondary ice production (squares) and after seeding (triangles). Temperature: −4°C. The plotted points are uncertain ±0.2 × 10^{-11} A from instrument noise.

persed throughout the chamber some 20 s after nucleation and continued to grow as moisture mixed into the cloud. The moisture supply was normally maintained throughout the experiment. The cold plate gave initial ice crystal concentrations (estimated from mean separation of crystals in the light beam) of ~1000 ml^{-1}; the other techniques both gave concentrations of ~100 ml^{-1}. Crystals began to sediment as their size reached 100–200 μm. The rime acquired positive charge in these experiments. When the larger concentrations of ice crystals were produced by seeding, no charge appeared until some 150–200 s had elapsed, after which the charging rate built up to a maximum, and then decreased to zero over a period of 50 to 100 s. This observation was consistent with the length of time taken for crystals to grow to 30–50 μm diameter and become large enough to be impacted by the rotating arm. For the lower crystal concentrations, charging always began within 10 s, the time required to grow crystals of this size. Visible water cloud disappeared ~30 s after seeding. In common with earlier observations no charge transfer was observed when the cloud consisted entirely of supercooled droplets. The effect of impact velocity on charge separation is shown in Fig. 5 for a plastic bubble seed. As crystals sedimented, cloud drops reformed and the charging rate fell to zero. At speeds below 0.7 m s^{-1} no detectable charge transfer took place. Charge transfer could be detected at 1.5 m s^{-1} and it increased rapidly with velocity to the maximum speed used, 3.5 m s^{-1} (Fig. 3, triangles). Assuming an overall collection efficiency of 0.5, Fig. 6 shows that the charge separated per crystal collision at −6°C increases rapidly with impact velocity, to a highest measured value of 5 × 10^{-16}C. This increase is signifi-

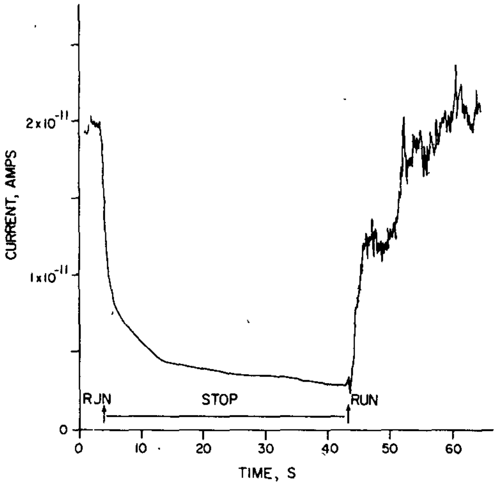

FIG. 4. Charging current resulting from secondary ice production as the riming rod is stopped and restarted. Temperature: −4°C.

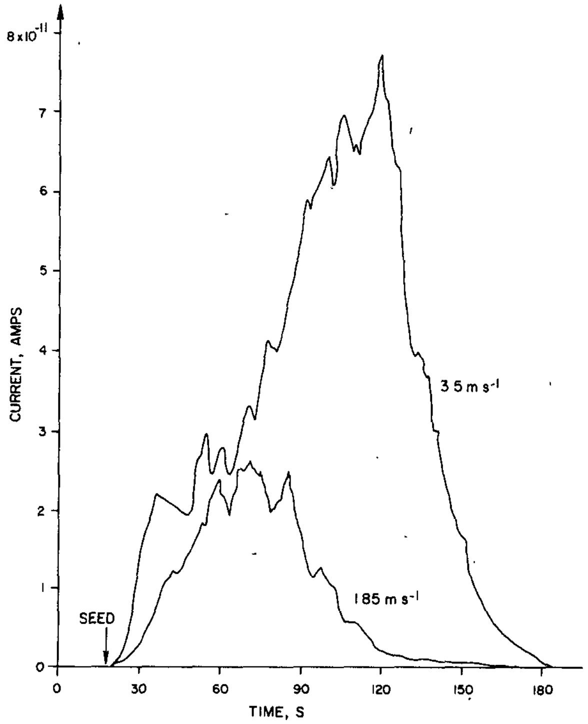

FIG. 5. Current to the riming rod due to the impact of ice crystals of diameter ⩾50 μm produced by seeding. Temperature: −5.5°C. Maximum charge per crystal collision: 5.3 × 10^{-16}C at 3.5 m s^{-1}, 2.5 × 10^{-16}C at 1.85 m s^{-1}.

cantly higher than is expected from a change of collision efficiency alone. The charge was somewhat less at both higher and lower temperatures, as shown in Fig. 7; this could be associated with the more extreme habit of the crystals as long columns near −4°C.

If it be assumed that the charge separation results only through ejection of secondary ice crystals, then a crude estimation, assuming that $^1/_{10}$ of the crystals are produced by the charging rod (the remainder being produced by the opposite rod, shielding cups and supporting arms), an instantaneous concentration of 1 crystal in 30 ml and a sedimentation velocity of 0.1 m s^{-1} gives a charge per secondary ice particle of 5×10^{-14}C. The interpretation that charge separation is occurring as the secondary ice particles are produced is consistent with the observation that charging begins almost instantaneously after the onset of rotation following a stop long enough to permit all ice particles to sediment as shown in Fig. 4. If collision were the dominant mechanism, a longer period—more than 10 s—would be required for the crystals to grow and their concentration to reach equilibrium. It follows that secondary ice crystals result in charge separation by two distinct processes—during production, maximum 5×10^{-14}C, and during collision, maximum 5×10^{-16}C per collision. In the present experiment the increase of charging rate which occurs ~100 s after the rotation is begun is to be attributed to the collision process. The difference of velocity dependence of secondary and seeded ice shown in Fig. 3 suggests that quite distinct physical mechanisms are responsible for charge separation.

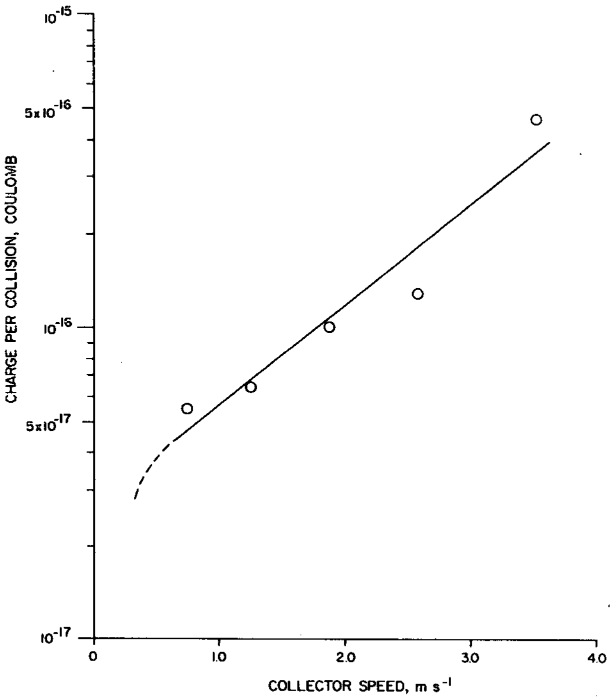

FIG. 6. Dependence of charge separation per crystal collision in impact with ice crystals produced by seeding. Temperature: −6°C.

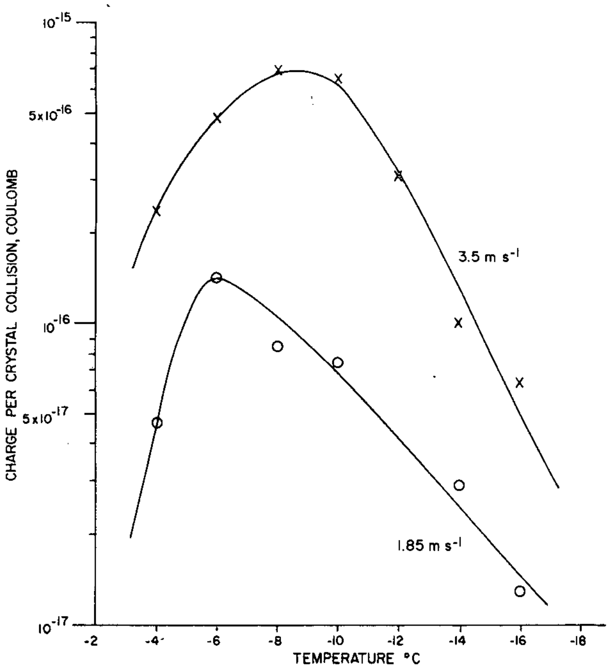

FIG. 7. Charge per collision as a function of temperature and collector velocity for ice crystals produced by seeding.

An important observation is that the sign of the charge acquired by the rime was positive both for artificially nucleated crystals and during multiplication charging. This is to be contrasted with the results of Reynolds *et al.* (1957) who obtained negative charging of the rime (10^{-14}C per collision), a result which has been used as the basis of a theory of thunderstorm electrification. An important difference in experimental technique between Reynolds *et al.* (1957) and the present work (private com-

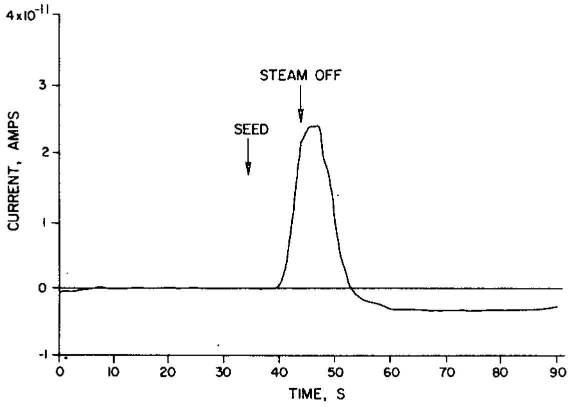

FIG. 8. Positive charging of the rod following seeding falls and reverses as the water vapor supply is cut off. Temperature: −6.5°C.

munication) is that they turned off the water vapor supply before making charge measurements, while in the present work the supply of moisture was maintained. In order to investigate this further, several experiments were conducted in which, after nucleation, the water source was turned off while charge transfer measurements continued. The sign of the rime charge started positive as in the previous experiments. As the saturation ratio decreased, and as the drops and vapor were depleted, the sign *reversed* to produce negative charging (Fig. 8). The present results are also to be compared with those of Takahashi (1978) who found that below −10°C, the sign of charge transfer during riming was dependent on temperature and liquid water content. At temperatures above −10°C, on the other hand, he observed positive rime charging, with no evidence of reversal for low liquid water content. Measurements of charge transfer in natural clouds have been made by Hobbs and Burrows (1966) who whirled an ice sphere through various types of cloud and snowfall. With both crystal aggregates and individual crystals, and in conditions close to ice saturation, they found negative charging of the sphere. There is some evidence from their work that the opposite sign of charge appeared when crystals collided in the presence of water drops.

In attempting to provide an interpretation of their results, earlier workers have invoked the sign and magnitude of the temperature differential between the ice target and the incident ice particle to provide a sign dependence of the separated charge; a target heated by radiation or by latent heat of freezing drops becoming negatively charged. The observations of Marshall *et al.* (1978), however, showing that radiant heating of an ice target gave an enhancement of charge transfer (target negative) independent of temperature difference >1°C, suggests that this criterion is far from adequate. Bearing in mind that any asymmetrical rubbing process must give significant local heating of the moving particle (Shio, 1978; Shio and Magono, 1971) as far as 0°C, it is unlikely that a small excess *temperature* of the target surface would, by itself, give rise to the observed effects. This leads to the conclusion that the surface character of the ice, as determined by the local saturation ratio, is the controlling factor. A similar saturation dependence has been found by Buser and Aufdermauer (1977) for ice crystals impacting on ice whose surface has been formed by deposition or sublimation at −45°C. The saturation ratio is necessarily related to temperature above or below ambient air temperature; we hypothesize that the sign of the charge transfer depends on frictional temperature excess of the incident ice particles in a surface layer whose character depends on the saturation ratio. Evidence of the temperature dependence of the depth of such a layer on ice comes from theoretical considerations (Fletcher, 1970) and from nuclear magnetic studies of Klividze *et al.* (1974). Anderson and Hallett (1979) have shown that the character of the layer must depend on supersaturation, as the nucleation and growth of ice layers in molecularly flat surfaces require a small but distinct critical supersaturation.

3. Implications

In applying these results to the atmosphere it is first necessary to assess conditions under which secondary ice crystal production can take place and the likely relative concentrations of graupel and vapor grown ice crystals which might result. Conditions for secondary ice production readily occur in clouds with warm bases (15–20°C) to give larger cloud drops (>25 μm diameter) by the time rising air reaches the −4°C level; they also occur in clouds with colder base temperatures (5°C), providing a maritime aerosol gives a smaller number of larger cloud drops (Hallett and Mossop, 1974a,b; Mossop, 1978a,b). The former situation occurs in Florida summertime convective thunderstorms, where, once secondary ice production begins, the number of graupel particles increases rapidly by the freezing of 0.1 to 1.0 mm diameter supercooled raindrops, present in concentrations of 1 ℓ^{-1}. This gives a rapid increase of secondary ice crystal concentration as these frozen drops rime in turn. This leads to graupel concentration as high as 30 ℓ^{-1} with vapor grown ice crystals concentration to 60 ℓ^{-1} (Hallett, *et al.*, 1978) in a period ~300 s. In this particular case, regions of charge separation would therefore be expected to be localized at sites associated with secondary ice production, and be carried either upward or downward with the air motion, charge separation continuing by the ice-graupel collision process. The field observations in Florida show quite definitely that this bimodal spectrum of ice particles exists—small vapor grown columns from 100 μm long occurring together with millimeter diameter graupel particles, at temperatures in the neighborhood of −12°C. The crystal sizes are consistent with their having grown from small secondary ice crystals found near the −4°C level. The laboratory results suggest that because of a saturation ratio dependence of the sign of charge transfer, graupel will become positively charged in an updraft due to both secondary ice crystal production and, to a lesser extent, to ice collisions. Negative charging of graupel, on the other hand, will occur through ice crystal collisions in downdrafts as the cloud drops evaporate and the saturation ratio nears ice equilibrium.

In clouds with colder bases (<5°C) the lack of coalescence leads to a quite different evolution of the ice phase, with higher cloud liquid water content at heights near the −12°C level than in the

Florida clouds (Christensen *et al.*, 1974). This raises the possibility of secondary ice formation from graupel growing with *surface* temperature at −4°C at these levels. The detail of the charge transfer taking place is dependent on the size and fall velocity spectrum of ice crystals, which is dependent in turn on the detail of the graupel and drop evolution. As a given convective cloud evolves, microphysical conditions lead to charge separation only in selected regions of the cloud volume, with charge separated by differential fall velocity of different sized ice particles, to be redistributed in the cloud in response to the overall mixing processes.

Acknowledgment. The authors acknowledge the support and encouragement given by Professor J. Latham during this project. The electronics were designed and built by W. Gaskell. This work was performed during a sabbatical leave of J. Hallett at UMIST, who is currently supported in this work by a Grant from the National Science Foundation, Atmospheric Sciences Section, Washington, DC.

REFERENCES

Anderson, B. J., and J. Hallett, 1979: Influence of environmental saturation and electric field on growth and evaporation of epitaxial ice crystals. *J. Crystal Growth*, **46**, 427–444.

Buser, O., and A. N. Aufdermauer, 1977: Electrification by collision of ice particles on ice or metal targets. *Electrical Processes in Atmospheres, Proc. Sixth Conf. Atmospheric Electricity*, H. Dolezalek, Ed., Reinhold, 294–301.

Choularton, T. W., J. Latham and B. J. Mason, 1978: A possible mechanism of ice splinter production during riming. *Nature*, **274**, 791–792.

Christensen, L., V. Keller and J. Hallett, 1974: Ice particle concentration in prehail convective cloud in Colorado. *Preprints Conf. Cloud Physics*, Tucson Amer. Meteor. Soc.

Church, C. R., 1966: The electrification of hail. Ph.D. thesis, University of Durham, U.K., $$$ pp.

Dye, J., and P. V. Hobbs, 1966: Effects of carbon dioxide on the shattering of freezing waterdrops. *Nature*, **209**, 464–466.

Fletcher, N. H., 1970: *The Chemical Physics of Ice*. Cambridge University Press, 271 pp.

Hallett, J., and S. C. Mossop, 1974a: The production of secondary ice particles during the riming process. *Nature*, **249**, 26–28.

——, and ——, 1974b: Secondary ice particle production by the riming process. *Preprints Conf. Cloud Physics*, Tucson, Amer. Meteor. Soc., 165–168.

——, R. I. Sax, D. Lamb and A. S. R. Murty, 1978: Aircraft measurements of ice in Florida cumuli. *Quart. J. Roy. Meteor. Soc.*, **104**, 631–651.

Hobbs, P. V., and D. A. Burrows, 1966: The electrification of an ice sphere moving through natural clouds. *J. Atmos. Sci.*, **23**, 757–763.

Kuettner, J., 1950: The electrical and meteorological conditions inside thunderclouds. *J. Meteor.*, **7**, 322–332.

Kvlividze, V. I., V. F. Kiselev, A. B. Kurzaev and L. A. Ushakova, 1974: The mobile water phase on ice surfaces. *Surface Sci.*, **44**, p. 60–68.

Latham, J., and C. D. Stow, 1969: Airborne studies of the electrical properties of large convective clouds. *Quart. J. Roy. Meteor. Soc.*, **95**, 486–500.

Marshall, B. J. P., J. Latham and C. P. R. Saunders, 1978: A laboratory study of charge transfer accompanying the collision of ice crystals with a simulated hailstone. *Quart. J. Roy. Meteor. Soc.*, **104**, 163–178.

Mossop, S. C., 1978a: The influence of drop size distribution on the production of secondary ice particles during graupel growth. *Quart. J. Roy. Meteor. Soc.*, **104**, 323–330.

——, 1978b: Some factors governing ice particle multiplication in cumulus clouds. *J. Atmos. Sci.*, **35**, 2033–2037.

——, and J. Hallett, 1975: Ice crystal concentration in cumulus clouds: Influence of the drop spectrum. *Science*, **186**, 632–634.

——, and E. R. Wishart, 1978: The mechanism for splintering during rime growth. *Geophys. Res. Lett.*, **5**, 1083–1085.

Reynolds, S. E., M. Brook and M. F. Gourley, 1957: Thunderstorm charge separation. *J. Meteor.*, **14**, 426–436.

Shio, H., 1978: Frictional electrification of single ice crystals. *J. Meteor. Soc. Japan*, Ser. II, **50**, 489–500.

——, and C. Magono, 1971: Frictional electrification of polycrystalline ice and single ice crystals. *J. Meteor. Soc. Japan*, Ser. II, **50**, 159–165.

Takahashi, T., 1978: Riming electrification as a charge generation mechanism in thunderstorms. *J. Atmos. Sci.*, **35**, 1536–1548.