Mixed-Phase Microphysics and Cloud Electrification

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ABSTRACT

A number of experimental studies have shown that sublimating ice acquires negative charge and ice undergoing vapor deposition acquires positive charge. Microphysical calculations are performed to determine the diffusional state (i.e., sublimation versus deposition) of riming graupel particles. Comparisons with earlier laboratory measurements of charge transfer to a rotating rimer in a cloud of supercooled water droplets and ice crystals again suggest that sublimating graupel particles charge negatively and graupel undergoing deposition charge positively. Implications for charge separation in thunderstorms are discussed.

1. Introduction

A considerable body of evidence has accumulated that indicates collisions between asimilar ice particles are responsible for the separation of electric charge in thunderstorms (Latham 1981; Illingworth 1985; Krehbiel 1986). Laboratory experiments on ice particle collisions show results (Reynolds et al. 1957; Takahashi 1978) consistent with large-scale observations of thundercloud electrical structure (Williams 1989).

Several recent laboratory studies (Baker et al. 1987; Avila et al. 1988; Dong and Hallett 1990; Rydock and Williams 1991) point up the importance of the growth condition of the ice surface in determining its charge polarity, irrespective of particle collisions. Baker et al. (1987) have advanced the interaction rule that "the fastest growing ice surface takes on positive charge."

This paper is concerned with microphysical calculations of the surface vapor condition of ice, comparisons with laboratory results on ice particle charging, and a reexamination of earlier laboratory experiments, all aimed at testing a generalization of the rule whose origin may be traced to Findeisen's work (1940): ice surfaces undergoing vapor deposition acquire positive charge (with or without a riming process), and surfaces undergoing sublimation acquire negative charge (with or without a riming process). The findings suggest that charge separation in thunderclouds is intimately linked with the mixed-phase microphysics of water substance, as embodied in the works of Wegener (1911), Bergeron (1935), Schumann (1938), and Ludlam (1951).

2. Physical boundaries on the diagram of liquid water content and cloud temperature

Laboratory work by Magono and Takahashi (1963), Takahashi (1978), Gaskell and Illingworth (1980), Jayaratne et al. (1983), and Keith and Saunders (1989) has greatly expanded the empirical information from the earliest experiment (Reynolds et al. 1957) on the conditions that give rise to charge transfer when (simulated) graupel pellets traverse cold regions of cloud containing supercooled water and ice crystals. Beginning with Magono and Takahashi (1963), the systematics of charge transfer have been represented in a diagram involving two fundamental parameters which empirically appear to control the charging process: the liquid water content (LWC) and the cloud environmental temperature (T). The purpose of this section is to include surface vapor conditions with the electrical information so that specific hypotheses for charge separation can be assessed. In subdividing the LWC–T diagram into specific regions, two boundaries shall be considered: the transition between wet and dry growth of graupel (Ludlam 1951), which was investigated earlier in the context of cloud electrification by Reynolds (1953), and the transition between sublimation and deposition of the graupel set up by the difference in saturation vapor pressures over liquid water and ice, respectively. Within the region of graupel sublimation on the LWC–T diagram, we shall also consider the effect of discrete droplets on the ice surface and the local zones of deposition that these droplets can create.

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The physics of wet and dry growth of graupel/hail is applied to the laboratory results of Takahashi (1978) with the equations set forth in Macklin and Payne (1967). In this continuum theory, the graupel particle with an assumed smooth spherical (or cylindrical) surface moves through a cloud with ambient temperature T_a and is uniformly bathed in supercooled water. The balance between heat released due to freezing and heat transferred to the environment by thermal and vapor transfer establishes a steady-state uniform temperature, T_g, for the graupel, which is systematically higher than T_a. The effective supercooled liquid water content, EW_c, associated with an equilibrium graupel temperature, T_g, is given in Macklin and Payne (1967) as

$$\mathrm{EW}_c V \frac{\{L_f + C_w(T_a - T_m) + C_i(T_m - T_g)\}}{\pi} = \chi \mathrm{Re}^{0.6} \frac{[\mathrm{Pr}^{1/3} K(T_g - T_a) + \mathrm{Sc}^{1/3} D_v L_v(\rho_g - \rho_a)]}{2R}. \quad (1)$$

Here R is the radius (R = 1.5 mm) of Takahashi's (1978) cylindrical rimer. The quantity χ is the numerical factor in the heat transfer coefficient which is chosen to be 0.28 based on Macklin and Payne (1967): for Takahashi's (1978) experiment the Reynolds number lies between 2100 and 2400 for a velocity V = 9 m s^{-1}; Pr and Sc are Prandtl and Schmidt numbers; K is the thermal conductivity of air; D_v is the vapor diffusion coefficient; L_v and L_f are the latent heats of vaporization and fusion; C_w and C_i are the heat capacities of liquid water and ice.

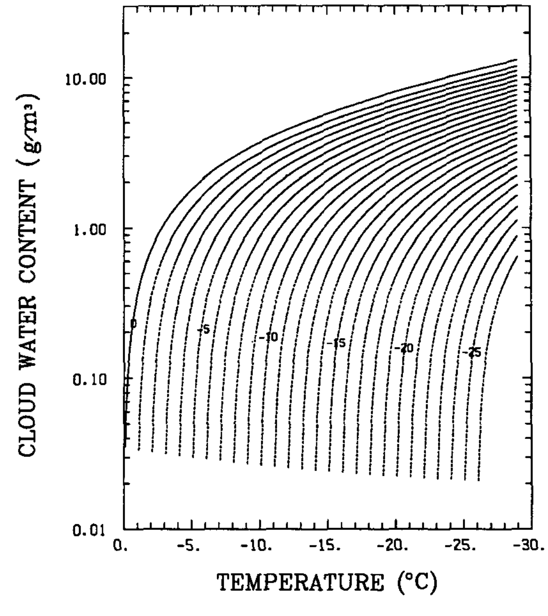

FIG. 1. Calculated isotherms of rimer (graupel) temperature, T_g, on the diagram of liquid water content and cloud temperature. The 0°C graupel isotherm (leftmost in the figure) marks the transition from dry to wet growth.

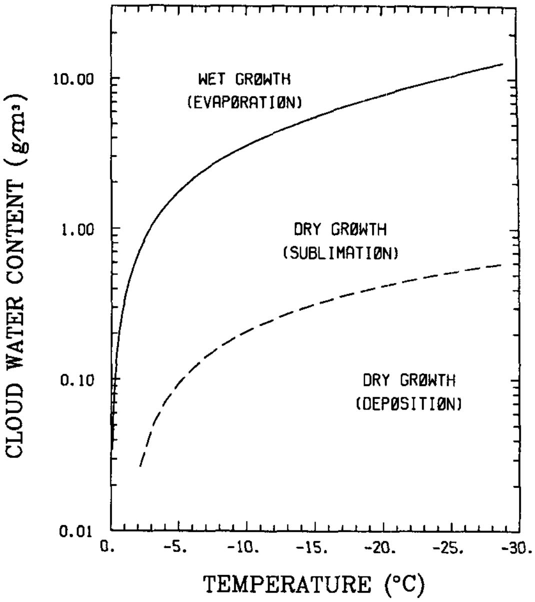

FIG. 2. Calculated boundaries between wet and dry growth by accretion and between sublimation and deposition for the rimer (graupel), applicable to the mixed-phase cloud in laboratory results of Takahashi (1978).

Figure 1 shows isotherms of graupel temperature, T_g, on the LWC–T diagram to illustrate the pronounced warmings at large values of liquid water content. The T_g = 0°C isotherm represents the boundary between wet and dry growth of the simulated graupel. For values of liquid water content larger than that needed for wet growth, the graupel surface is in a liquid state and is undergoing evaporation, while experiencing net growth by accretion. At smaller values of liquid water content, the T_g isotherms are increasingly parallel to the vertical axis but slightly offset from the cloud temperature since the rimer remains warmer than the environment even at low LWC.

The higher temperature of graupel relative to its environment (for $T_g < 0$°C) will promote sublimation of ice if the surface temperature, T_g, is sufficiently large relative to the environment at T_a. Here again the particle is losing mass by sublimation but gaining mass by accretion. When the equilibrium vapor pressure of ice at the temperature T_g falls below the equilibrium vapor pressure of the environmental supercooled water at T_a, the vapor transfer for the graupel shifts from sublimation to deposition. This lower boundary on the LWC–T diagram, shown in Fig. 2, is readily calculated using the T_g values in Fig. 1 and the Clausius–Clapeyron relation. In the lower region, the graupel particle is gaining mass by both vapor deposition and by accretion.

As Macklin and Payne (1967) have noted, the continuum theory used above to determine equilibrium graupel temperatures and vapor state ignores the discrete nature of individual droplets arriving at the graupel surface. Instead of warming monotonically from T_a to T_g, such droplets instead are quickly warmed by the local release of the latent heat of fusion to 0°C and remain at this temperature for a finite time τ_{af} before completely freezing and then cooling to T_g. Baker et al. (1987) have noted that the transient liquid droplet acts as a source of water vapor, which subjects the droplet's graupel substrate to vapor deposition in the droplet's vicinity. We make use of the calculations presented by Baker et al. (1987) to investigate the competition between deposition and sublimation in the sublimation region of Fig. 2 caused by the discrete arrival of supercooled droplets.

In Baker et al. (1987), sublimation of the graupel surface is impossible since $T_g = T_a$ by assumption. By relaxing this condition and allowing the graupel to warm by accretion according to the continuum theory (the results in Fig. 1), we can equate the deep-field sublimation vapor theory [Eq. (12) in Baker et al.] with the local depositional flux [Eq. (22)] caused by the transient liquid droplet to determine the size of the zone around the droplet in which deposition dominates over sublimation. The radius of this zone

$$r^* = \left[\frac{3a^2R}{2}\frac{\rho_w(0°\mathrm{C}) - \rho_I(T_g)}{(\rho_I(T_g) - \rho_w(T_a))}\right]^{1/3} \qquad (2)$$

where a is the droplet radius, R is the graupel radius, ρ_w is the equilibrium vapor pressure of liquid water, and ρ_I the equilibrium vapor pressure of ice. For values again representative of Takahashi's (1978) experiment, $a = 5$ μm and $R = 1.5$ mm. The area of the circular zone of deposition, πr^{*2}, around each droplet on the graupel surface in the surrounding field of sublimation is two to three orders of magnitude larger than the cross-sectional area of the droplet.

For a monodisperse population of supercooled droplets with radius a, the droplet concentration is related to the cloud water content W_c by

$$N\rho\frac{4}{3}\pi a^3 = W_c. \qquad (3)$$

A simple statistical argument shows that the fractional area of sublimating graupel surface that is undergoing deposition by virtue of local droplets, each with a lifetime before freezing of τ_{af} [given approximately by Eq. (36) in Baker et al. 1987], is the nondimensional quantity

$$N \cdot V \cdot \pi r^{*2} \cdot \tau_{af}. \qquad (4)$$

Numerical calculations (which make use of the T_g distribution shown in Fig. 1) show that this quantity is substantially less than unity within the sublimation region of the LWC–T diagram. As expected, the fractional area undergoing deposition vanishes at the wet growth boundary in Fig. 2 (where deposition ceases) and grows to 100% as the lower deposition–sublimation boundary is approached (where sublimation ceases). Since the fractional area undergoing deposition never exceeds a value of a few percent over most of the intermediate area in Fig. 2, we conclude on the basis of this simple model that deposition associated with discrete droplets on the graupel surface does not strongly erode the field of sublimation predicted by the continuum theory, at least on an areal basis.

Comparisons between the cloud microphysical predictions for the LWC–T diagram and laboratory results for the polarity of graupel charging from Takahashi (1978) are shown in Fig. 3. While many laboratory experiments of this nature have been performed (Reynolds et al. 1957; Gaskell and Illingworth 1980; Jayaratne et al. 1983; Baker et al. 1987; Keith and Saunders 1989), the results of Takahashi most thoroughly explore the LWC–T parameter space and in this sense are most suitable for comparison with the theoretical results. Only Takahashi (1978) explores the wet growth regime. It is fair to note, however, that significant discrepancies in the parameter space plots of laboratory results have been identified (Jayaratne et al. 1983; Saunders and Zhang 1987, Fig. 5; Williams 1989, Fig. 2) and will be discussed in a later section. When judged against a large number of field observations (Williams 1989) of the electrical structure of clouds, Takahashi's lab results show the best agreement, as will be further discussed in section 6. The comparisons with Takahashi in Fig. 3 indicate that the negative charging of graupel (black circles) is reasonably well correlated with the zone of sublimation in dry growth, at least for temperatures lower than −10°C. For $T > -10$°C, the rimer charging is positive and the earlier

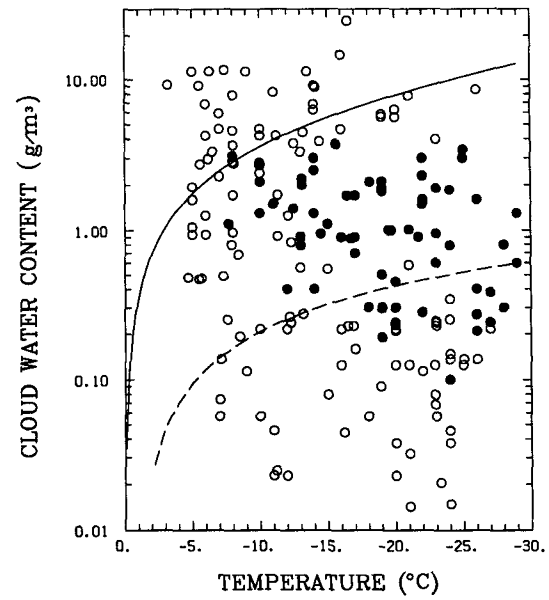

FIG. 3. Comparison between laboratory results of Takahashi (1978) and the theoretical results based on mixed-phase microphysics from Fig. 2. Black (white) dots denote negative (positive) charge transfer to the rimer (graupel).

consideration of local zones of deposition [Eq. (2)–(4)] does not appear to account for the discrepancy in sign. This result is inconsistent with the sublimation/deposition hypothesis and this remains a puzzle in this study. Deposition, in the lower part of the LWC–T diagram, is associated with positive charging of graupel (open circles), though the slope and location of the predicted boundary shows considerable discrepancy with the data. At large values of LWC in the upper part of the diagram the graupel surfaces are in wet growth and acquire positive charge in ice crystal collisions, though some open circles fall within the dry growth region. The inferred region of wet growth is consistent with Takahashi's (1978) analysis of the rimer surface, which indicated a wet surface for experimental runs in this region of the diagram. In fact, Takahashi identified empirically all three microphysical regimes of the LWC–T diagram which this paper has identified theoretically in Fig. 2. The three regimes illustrated in Fig. 11 of Takahashi (1978) correspond to a) deposition in dry growth, b) sublimation of ice in dry growth, and c) evaporation of liquid in wet growth, according to the calculations already discussed. (The designations "high" and "low" liquid water content in Takahashi's Fig. 11 are incorrectly reversed.) Specific experimental runs discussed by Takahashi for each of these three regimes are represented by points in the LWC–T diagram of Fig. 2: (a) −22°C, 0.1 g m^{-3}, (b) −22°C, 0.9 g m^{-3}, and (c) −14°C, 5.6 g m^{-3}, respectively.

3. Comparisons with other laboratory results

The association of negative charging with sublimation and positive charging with deposition is consistent with several earlier experimental results. Studies of the separation of charge from surfaces of ice undergoing both vapor deposition and sublimation (and in the presence of riming) are summarized in Table 1.

Findeisen (1940) and Findeisen and Findeisen (1943) exposed cold objects (−70°C) to streams of dry and humidified air. A predominance of negatively charged fragments of ice was ejected from the cold surface during deposition and frost growth; positively charged fragments were seen when the frost was exposed to a dry airstream and began to sublimate. In these studies the ice accumulation was referred to as "rime," but this was not confirmed by direct observation and it may well have been frost alone.

Latham (1963) exposed ice surfaces to airstreams that were either colder or warmer than the ice but in which the vapor content was not a controlled parameter. The results (Table 1) were interpreted in light of the thermoelectric effect, but a possible alternative explanation, and one more in line with other results in this paper, is that the warmer airstream subjected the ice to deposition (and positive charging), while the colder airstream induced sublimation (and negative charging). This interpretation is partly supported by subsequent experiments (Latham and Stow 1965) in

TABLE 1. Summary of experiments on charging of ice surfaces (in the absence of particle collisions).

Investigator	Study	Principal results
Findeisen (1940)	Ventilated cold surfaces	+ charge acquisition during deposition − charge acquisition during sublimation
Findeisen and Findeisen (1943)	Ventilated cold surfaces	+ charge acquisition during deposition − charge acquisition during sublimation
Clay and Kramer (1947)	Ventilated cold surfaces	+ charge acquisition for warm airstream + charge acquisition during deposition (initially only)
Latham (1963)	Surface frost exposed to airstream	+ charge acquisition for airstream warmer than deposit − charge acquisition for airstream colder than deposit
Latham and Stow (1965)	Ice surface in cold stream of nitrogen gas	− charge acquisition during sublimation
Schaefer and Cheng (1971)	Cold object ventilated by buoyancy-induced airflow	+ charge acquisition during deposition
Takahashi (1973)	Electrification of ice crystals	+ charge acquisition during deposition − charge acquisition during sublimation
Buser and Aufdermaur (1977)	Ice specimen at −45°C	+ charge by surface formed by deposition − charge by surface formed by sublimation
Dong and Hallett (1990)	Rime undergoing vapor diffusion in conditions of controlled saturation	+ charge acquisition during deposition − charge acquisition during sublimation
Rydock and Williams (1991)	Cold object ventilated by buoyancy-induced airflow	+ charge acquisition during deposition in large vapor gradient

which a dry nitrogen airstream was used, thereby guaranteeing sublimation of the ice, and systematic negative charging of the ice was observed.

Schaefer and Cheng (1971) investigated the growth of an ice surface and the ejection of charged fragments therefrom on an ice sphere (at $T = -20°C$) in buoyancy-driven air flow. Consistent with Findeisen's (1940) findings, the cold object (sphere) acquired positive charge as a result of the predominance of negative charge on the ejected ice fragments. In these experiments, the buoyancy-driven flow is observed to contain a sheath of supercooled water adjacent to the growing ice surface.

Rydock and Williams (1991) pursued this charging phenomenon to explore its dependence on imposed temperature and vapor gradients. Over a wide range of conditions in which growth by vapor deposition occurred, the same polarity of charge was found: negative charge in the ejected fragments with positive charge left on the growing ice surface. Rydock and Williams (1991) suggest a link between the latter phenomenon and the experiments involving ice particle collisions; namely, that the polarity of charged ice fragments ejected from a surface undergoing deposition (or alternatively sublimation) is identical with the polarity of charge available for transfer from the ice surface when a smaller ice particle collides with it.

It is important to emphasize that the temperature and vapor gradients present in experiments by Findeisen (1940), Findeisen and Findeisen (1943), Schaefer and Cheng (1971), and Rydock and Williams (1991) are at least an order of magnitude greater than gradients present at the surfaces of ice particles in thunderclouds. Dong and Hallett (1990) have recently reported on laboratory measurements of charge transfer to and from a rimed surface in a diffusion chamber under conditions more representative of the atmosphere. Consistent with other results just discussed, they find positive charge acquisition associated with deposition and negative charging associated with sublimation, though the agent of charge transfer was not identified.

Marshall et al. (1978), Gaskell and Illingworth (1980), Baker et al. (1987), and Avila et al. (1988) have described experiments whose results may also provide important links between the earlier studies of vapor-exchanging ice surfaces (Table 1) and the experiments involving ice particle collisions in a riming environment to be discussed in the next section. By controlled cooling (heating) of their graupel target by a few degrees relative to its environment, these studies have shown that collisions with smaller ice particles result in the systematic acquisition of positive (negative) charge by the target. These results again show positive charge acquisition linked with depositional growth and negative charge acquisition linked with sublimation and suggest that one underlying phenomenon may be responsible for these many experimental results.

4. Comparisons of laboratory simulations of the thunderstorm mixed-phase region

The recognition that charge separation in thunderclouds was taking place in the cold part of the cloud has led to many laboratory experiments that attempt to simulate mixed-phase conditions (Reynolds et al. 1957; Magono and Takahashi 1963; Takahashi 1978; Hallett and Saunders 1979; Gaskell and Illingworth 1980; Jayaratne et al. 1983; Baker et al. 1987). As noted earlier, the results best suited for comparisons with our results are those of Takahashi (1978), and fortunately, these results (Fig. 3) are quite consistent with the original findings of Reynolds et al. (1957): "In general, the sign of the charge on the rimer is positive when the crystals are numerous in relation to the droplets, and negative when the reverse situation obtains and the liquid-water content is relatively high." The predominant control of the charge polarity by liquid water content is readily apparent in Fig. 3. Magono and Takahashi (1963) show results over a smaller portion of the LWC–T diagram than shown by Takahashi (1978), but which are consistent with the latter results.

Other results show less consistency. Gaskell and Illingworth (1980) find a charge reversal temperature (at fixed LWC) which is close to the $-10°C$ value found by Takahashi (1978), but do not find a transition to positive rimer charge at low values of LWC. The charge reversal temperatures found by Jayaratne et al. (1983) and Baker et al. (1987) (in the same laboratory) are still lower ($-17°$ to $-20°C$) than that of Takahashi (1978), and positive rimer charge is detected at high LWC over a wide range of temperature, in direct contradiction with the results of Reynolds et al. (1957), Magono and Takahashi (1963), and Takahashi (1978). The reasons for these differences have not been adequately identified although Saunders et al. (1985) emphasize the role of differences in rimer velocity in the various experiments. Reynolds et al. (1957) and Takahashi (1978) both operated with a rimer velocity of 9 m s^{-1}, whereas Jayaratne et al. (1983) used 3 m s^{-1}. The riming studies of Macklin (1962) and Zhang and Saunders (1987) suggest that the rime density in Takahashi (1978) may be two to three times greater than that in Jayaratne et al. (1983) because of the velocity difference alone. The low-density rime with a more uneven surface is expected to conform less well to the Schumann–Ludlam theory used here for Takahashi's results. We recognize that microphysical conditions in thunderclouds are highly variable and that one must be cautious in extrapolating laboratory results to thundercloud conditions. While it is conceivable that none of the laboratory simulations is relevant to thunderclouds, we find that the results of Takahashi (1978) shown in Fig. 3 are consistent with many of the large-scale observations of thunderclouds and shall present evidence for this claim in section 6.

5. Hypotheses for charge polarity in ice particle collisions

The calculations presented in section 2 and their comparison with laboratory results permit a reexamination of various hypotheses for thundercloud charge separation. Among the many hypotheses that have been suggested in the last 50 years for the selective transfer of charge when small ice particles collide with graupel/hail particles in a supercooled cloud are the following.

H1: Graupel/hail in wet growth charge negatively following the Workman–Reynolds effect (Workman and Reynolds 1950)

H2: The warmer ice particles charge negatively; colder particles positively (Reynolds et al. 1957; Brook 1958; Takahashi 1978)

H3: The faster-growing particles (by vapor transfer) acquire positive charge (Baker et al. 1987)

H4: Sublimating graupel charge negatively; graupel undergoing deposition charge positively (Gaskell and Illingworth 1980; Jayaratne et al. 1983; this paper).

We will now consider the evidence for and against these various hypotheses.

First, H1 is not supported by the comparisons in Fig. 3, which indicate that graupel particles in the wet growth regime take on positive charge in collisions with supercooled water droplets and ice crystals. This discrepancy was pointed out earlier by Magono and Takahashi (1963). The observation of positive rimer charge is consistent, however, with other systematic results on the polarity of charge acquired by ice in contact with liquid water (Sohncke 1886; Fairbrother and Wormell 1928).

Second, H2 is only partly supported by the results shown in Fig. 3. Within the dry growth regime in the laboratory cloud at water saturation, the simulated graupel particles growing by riming are systematically warmer than the surrounding ice crystals (growing by deposition), for all values of the cloud water content. This hypothesis correctly accounts for the observed and calculated acquisition of negative charge by the rimer at high LWC in Fig. 3, but does not account for the observed and calculated transition to positive rimer charge at lower values of LWC. Of course, in a real thundercloud, because of the decreasing cloud temperature with height and the possible descent of the graupel at several meters per second relative to the smaller ice particles, it is possible for the graupel to be colder than the adjacent smaller particles because of thermal inertia. This situation cannot occur in the laboratory setup, however, and so is not examined further here.

Reynolds et al. (1957) performed an auxiliary experiment with an infrared lamp as a further test of H2. They reported that "when the cloud conditions were adapted for strong positive charging of the rime (when the cloud was constituted principally of ice crystals), and when the rime was heated, strong negative charging promptly resulted." If the lamp's heat affects T_g alone (and not T_a), then this experiment can be illustrated in Fig. 2 as an excursion at fixed T_a across T_g isotherms (Fig. 1) from the positive to the negative region (Fig. 3). The interpretation of this experiment by Reynolds et al. (1957) and later by Brook (1965) presupposes that the unilluminated rimer is colder than the surrounding ice crystals, a questionable assumption. We believe that the positive-to-negative transition in their experiment lends more support to H4 than to H2, as illustrated in Fig. 3.

Baker et al. (1987) proposed H3 to account for positive charging of the rimer, a consistent finding in the Manchester laboratory (Hallett and Saunders 1979; Jayaratne et al. 1983; Baker et al. 1987; Keith and Saunders 1989). Calculations presented in section 2 suggest that the deposition effects associated with individual droplets at 0°C on the colder graupel surface will not suppress the larger-scale sublimation, which dominates in the central region of Fig. 2. As schematically illustrated in Fig. 4, H3 correctly accounts for comparisons only in the sublimation portion of the dry growth region in Fig. 3. If sublimation of ice is treated as negative growth, the graupel particles are growing less fast (by vapor transfer) than the ice crystals. However, while the graupel particles begin to grow by vapor deposition in the lowest region in Fig. 2, they will not be growing *faster* than the ice crystals in this region and so will not account for observed and calculated positive rimer charge there.

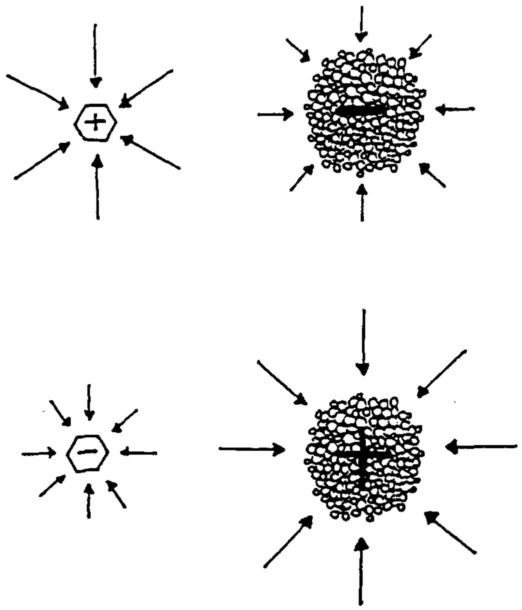

FIG. 4. Schematic illustration of tripole formation according to hypotheses (H3): the faster-growing particles acquire positive charge.

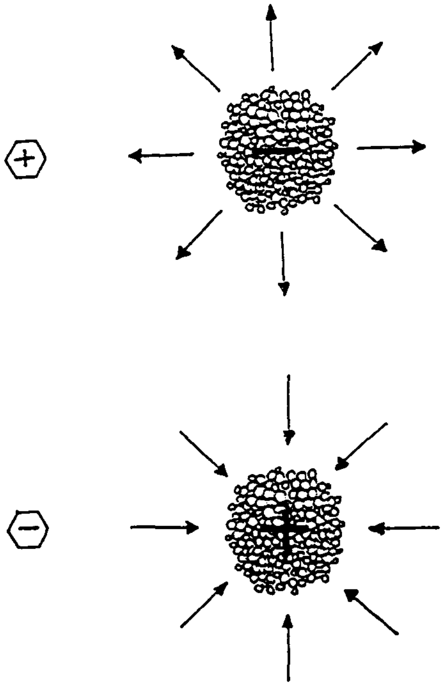

FIG. 5. Schematic illustration of tripole generation according to hypothesis (H4): sublimating graupel particles take on negative charge; graupel particles undergoing deposition take on positive charge.

The hypothesis advanced in this paper, H4, has its origins in the studies of Findeisen (1940). This hypothesis correctly accounts for the polarity of charge taken on by an ice surface in experiments (reviewed in section 3) with or without riming but with no collisions (Table 1), and in experiments without riming but with particle collisions (Marshall et al. 1978; Gaskell and Illingworth 1980; Baker et al. 1987; Avila et al. 1988). Hypothesis H4 is schematically illustrated in Fig. 5 for comparison with the subtly different H3. Unlike H3, H4 gives special attention to the graupel (rimer). This hypothesis correctly accounts for the results in Fig. 3 showing negative rimer charge in the sublimation region and positive rimer charge in the deposition region of dry particle growth, even when the temperature, which controls the sublimation–deposition transition, is modulated by an active riming process.

The validity of H4, supported by the laboratory results of Takahashi (1978), has been called into question when applied to experiments involving riming and collisions by both Gaskell and Illingworth (1980) and by Jayaratne et al. (1983) on the basis of their own experimental results. In the latter study, this discrepancy is stated clearly: ". . . The parameter controlling the sign of the charge transfer is the surface state of the target which charges negatively when evaporating (sublimating) and positively when growing. This effect was completely masked while riming took place simultaneously. . . ." Again, we do not understand the reason for the discrepancy, but note that the distinction between sublimation and deposition for low-density graupel is probably not well specified by Schumann–Ludlam theory. We are inclined to believe in the validity of Takahashi's (1978) results because of the consistency they bring to a large number of experimental results under H4 and because of their consistency with cloud-scale observations (section 6).

The comparisons discussed thus far have been concerned mainly with the polarity of charge transfer. When magnitude is considered in the collision experiments, virtually all laboratory results are in agreement that the charge transfer is substantially larger in the case of riming than in the case of vapor transfer alone. This circumstance again raises the question of the specific role of the riming process and what actually happens when an ice particle collides with a rimed or an unrimed graupel surface. It is well recognized that rimed graupel surfaces are delicate and fragile (Vardiman 1978; Vali 1980) and in fact depart significantly from the smooth spherical surface assumed in the derivation of (1). The velocity and crystal size dependence of collisional charge transfer in the rimed case (Keith and Saunders 1989) suggest the possibility that the ice crystal kinetic energy is a fundamental parameter. Simple calculations using a surface energy for ice of 2×10^{-2} J m^{-2} show that this kinetic energy is sufficient to fracture rime branches on the graupel surface and play a role in charge transfer. This idea, together with the documented support for H4, suggests that temperature gradients at the irregular surfaces of graupel particles undergoing vapor transfer be more thoroughly investigated as a source for charge transfer in collisions. Such temperature gradients are ignored in the continuum theory, but in the presence of highly localized latent heat transfers in riming, sublimation, and deposition, the local temperature differences can be substantially greater than the particle-to-particle temperature differences considered in H2. The sign of these temperature gradients will of course correlate with the state of surface growth.

6. Constraints imposed by large-scale observations

The microphysical conditions within thunderclouds are well recognized to be highly variable and comparisons with laboratory simulations must be treated cautiously. However, the laboratory results in Fig. 3 show considerable agreement with large-scale observations when both cloud water content and electrostatic structure are considered.

For comparison with the results in Fig. 3, the adiabatic LWC in the mixed-phase region of thunderstorms at midlatitude lies in the range of 4–6 g m^{-3} (see Fig. 3.9 in Ludlam 1980). While occasionally superadiabatic values are detected, and frequently peak values attain the adiabatic prediction, the most prevalent values are 20%–30% of the adiabatic values (e.g., Musil and Smith 1989). With these typical LWC values of 1–2 g m^{-3}, Takahashi's (1978) results suggest dry growth microphysics and sublimating negatively charged graupel over a wide range of ambient cloud temperature. The theoretical calculations in Fig. 2 show that the sublimation/negative graupel region extends over at least an order of magnitude in LWC over this range of temperature, lending considerable robustness to the phenomenon, and is consistent with the prevalence of negative charge below positive charge in thunderclouds.

As far as the large-scale electrical structure of thunderclouds is concerned, observations (Williams 1989) continue to show the main negative charge center at a range of heights consistent with Takahashi's charge reversal temperature ($T_a = -10°C$), and a positive dipole length of several kilometers, consistent with the selective transfer of negative charge to graupel over a substantial range of height (and cloud temperature). Findings in recent field studies (Dye et al. 1988; Ziegler et al. 1991) also support the applicability of Takahashi's (1978) laboratory data.

Electrical observations (Williams 1989) also show a substantial tapering off of negative charge beneath the center of main negative charge, suggesting that the inferred negatively charged graupel are unloading their negative charge and taking on positive charge. Figure 3 suggests that the observed acquisition of positive charge may occur by a mechanism not accounted for by H4 and our calculations based on classical microphysics, or alternatively by a transition (at fixed LWC) from the dry growth to the wet growth regime. The latter possibility was first suggested by Magono and Takahashi (1963). Graupel particles falling through the updraft between the particle balance level (Lhermitte and Williams 1985) and the melting level should be candidates for positive charge acquisition. Observations are needed to see if positively charged graupel above the 0°C isotherm are experiencing transient wet growth.

It does not seem likely that the deposition/positive rimer regime in Fig. 3 is responsible for the lower positive charge in thunderclouds (as Fig. 5 suggests) because the liquid water contents are so small. This region of the parameter space may have applicability, however, in the stratiform precipitation of mesoscale convective complexes, where dominant lower positive charge has been inferred to exist.

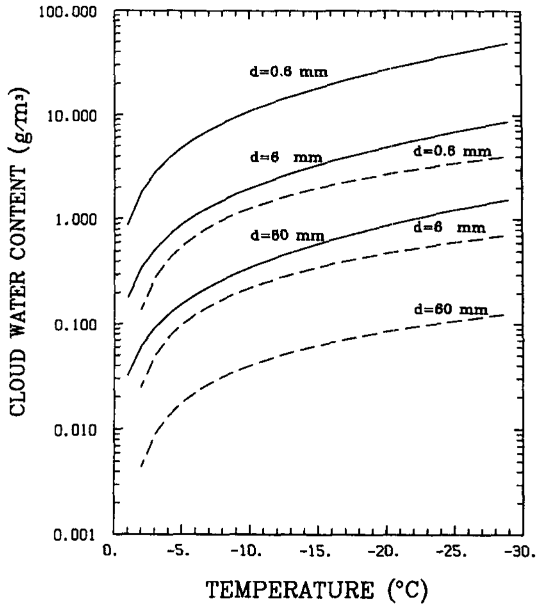

FIG. 6. Calculated boundaries between wet and dry growth (solid lines) and between sublimation and deposition (dashed lines) for spherical ice particles, for three values of particle diameter.

Additional microphysical calculations have been performed with an equation similar to (1) but appropriate for spherical graupel/hail (Bailey and Macklin 1968) and with velocities given in Pruppacher and Klett (1980, p. 345) and a graupel density of 0.5 g cm^{-3}. These results are shown in Fig. 6 for three graupel diameters (0.6 mm, 6 mm, and 60 mm). The solid curves represent the respective wet–dry growth transition and the dashed curves represent the respective boundaries between sublimation and deposition in dry growth. The results in Fig. 6 suggest that graupel particles in the important range of $d = 1$ mm to $d = 1$ cm will find themselves in the sublimation/negative polarity regime for a wide range of ambient temperatures for representative liquid water contents discussed above.

The results in Fig. 6 also predict that ice particles with diameters of 10 mm and larger (e.g., hailstones) will find themselves in wet growth for representative cloud liquid water contents, and hence should acquire positive charge. The behavior may help to explain the clustered cloud-to-ground lightning activity of positive polarity observed beneath extraordinarily vigorous hail-producing storms at midlatitude. Techniques are needed to distinguish wet from dry growth in the in situ measurements of precipitation particle charge, and to check the expectations based on Takahashi's (1978) laboratory measurements.

In light of the evidence for charge reversal associated with the transition form dry to wet growth (Fig. 3), new emphasis should be placed on developing polarization-diversity radar techniques for identifying the growth regime of graupel particles.

7. Conclusion

The results of these calculations and our review of laboratory and field observations support the earlier claim (Reynolds et al. 1957) that charge separation in thunderstorms is intimately related to classical mixed-phase microphysics, but the hypothesis advanced in the latter study for the polarity of charge transfer is not supported. The hypothesis that sublimating graupel acquire negative charge and graupel undergoing deposition acquire positive charge is most strongly supported by the calculations and comparisons. This hypothesis is supported by Takahashi's (1978) results in riming conditions, and by many earlier experiments in which riming played no role. Further understanding of the underlying physical mechanism requires a molecular characterization of ice surfaces undergoing both sublimation and deposition and a description of the ice particle–graupel collision at subparticle scales.

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