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# Charge transfer in convective thunderclouds induced by molecular interface crossing and free energy reduction

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## Abstract

In spite of many experiments showing that charge is transferred during collisions of cloud particles, the precise mechanism of the charge separation and its effect on the spatial charge distribution in thunderclouds is not yet understood. Collision processes between two cloud particles could be treated as composed of three phases: before contact, contact and withdrawal. Interfacial mass and charge transfer between proximal and contacting water or ice surfaces depends on the balance of competing intermolecular interactions at the interfaces. This paper attempts to demonstrate how the Free Energy Reduction by Molecular Interface Crossing (FERMIC) model could be complementarily applied and fully implemented into existing concepts to interpret complex atmospheric charge transfer processes, offering a novel qualitative physical explanation to the experimental results of charge separation in thunderclouds assumed to occur before contact. The FERMIC process has been extensively applied to the understanding of the transfer of mass or charge across liquid interfaces, self-organized molecular assembly in physical and biological processes and in the development of biomaterials. However, it was never used to explain the interfacial mass and charge transfer processes occurring during collisions of hydrometeors in thunderclouds where strong electrification and lightning are common. © 2001 Elsevier Science B.V. All rights reserved.

*Keywords:* Thunderstorm charging; Liquid-like layer; Graupel; Ice crystal; FERMIC

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## 1. Introduction

The generally accepted charging mechanism in thunderclouds is primarily based on collisions between large falling (with respect to the ascending air) hail particles (or graupel pellets) and small ascending ice crystals in the presence of water droplets (Williams, 1988; Dash et al., 1995). According to a recent theory, the charge transfer during collisions of ice particles is associated with the transfer of microscopic quantities of water at their surfaces (Baker and Dash, 1989, 1994). The existence of liquid-like water layers on the surface of ice at temperatures far below the bulk melting temperature has been verified experimentally by Holden (1998). Laboratory experiments show that both the magnitude and the sign of the separated charge depend on cloud temperature and cloud liquid water content (LWC) (Takahashi, 1978; Jayaratne et al., 1983). Observations in clouds suggest that the strongest electrification processes occur at temperatures around −15°C (Williams, 1988). Interpretations of atomic force microscope measurements, AFM, (see Döppenschmidt and Butt, 2000), indicate the existence of a 12-nm thick water layer on ice at −15°C. Remarkably, the thickness of this liquid-like layer decreased with temperature, from 32 nm at −1°C to 1 nm at −38.5°C, suggesting that the charge transferred in the cloud could depend on the thickness of the liquid-like layer on the ice crystal surface and be proportional to the number of the transferable molecules (and ions), accounting for the observation that there is practically no charging at temperatures below −38°C.

While it is generally accepted that collisions between cloud particles are the main cause for thundercloud electrification (Wettlaufer and Dash, 2000), only very little is known about the mechanism responsible for the transfer of charge (Vonnegut, 1995). If indeed, charge is transferred when microscopic quantities of water are transferred across the interface, charge exchange must depend on the contact time and the contact area. The values of these parameters during collisions between small ice crystals and large hail pellets are only poorly known. Measurement of contact time and estimation of contact area are in no way trivial and the meager information we have mainly comes from laboratory experiments, which may be quite different from the actual values in the clouds. Practically, no information is available for the transfer processes occurring before the contact.

Laboratory experiments (Levin and Hobbs, 1971; Levin and Machnes, 1977) show that as two water drops approach each other, a layer of compressed air develops between them, slowing their approach and on occasion preventing coalescence. Let us assume that a similar air barrier is formed at the contact zone of two approaching ice particles. If these ice particles each has a water layer (or quasi-liquid layer) on its surface, the Free Energy Reduction by Molecular Interface Crossing (FERMIC) process presented below predicts that surface material from these liquid-like layers will be transferred before the interfacial contact, in a preferential direction determined by thermodynamic stability criteria. In view of the considerable kinetic energy of the hydrometeors and the effects of turbulence on their translational and rotational motion, the interfacial area of interaction prior to the collision, the contact zones and the interfacial separation zones following the collision of the particles, may not be the same. This makes it possible to transfer material and charge between the colliding particles during the three stages or phases of the interaction. In the present paper we analyse the first phase: for charge to be separated in the cloud during this phase, we suggest that water molecules are transferred together with ions across the interface of the approaching curved surfaces by the Free Energy Reduction by Molecular Interface Crossing (FERMIC) mechanism, (Sommer et al., 1999).

## 2. The FERMIC model

For liquids in interfacial proximity, it was possible to derive a mechanism for pumping molecules from small droplets to larger ones solely from first principles. From the van der Waals model describing the intermolecular interaction between adjacent liquid molecules, the maximum molecular instability (fluctuations, most probable transfer of molecules) between two approaching liquid surfaces of unequal curvature is expected to occur at the area near the point of contact. In the simplest formulation of the van der Waals model, the interaction energy of a molecule of the bulk liquid phase is given by:

$$E_\mathrm{b} = \frac{1}{2} z_\mathrm{b} W_\mathrm{AA}, \tag{1}$$

where $W_\mathrm{AA}$ represents the pair interaction energy. The corresponding expression for molecules at the plane liquid surface is given by:

$$E_\mathrm{s} = \frac{1}{2} z_\mathrm{s} W_\mathrm{AA}, \tag{2}$$

where $z_\mathrm{s}$ is the coordination number of a surface molecule. The ratio between the energies $E_\mathrm{b}$ and $E_\mathrm{s}$ is approximated by the effective part of the ratio $z_\mathrm{b}/z_\mathrm{s} \geq 1/2$. The physical meaning of this ratio is that a surface molecule is attracted anisotropically and by a lesser number of closest neighbors than in the interior of the liquid. The two-dimensional model is often applied in the calculation of the total intermolecular interactions and the quantitative estimation of surface tensions in liquids (Evans and Wennerström, 1994; Huyskens and Luck, 1991). Going over to curved liquid surfaces, it can be easily seen that the coordination number of a surface molecule decreases as the curvature of the surface increases. From a 3D model of a spherical drop (or ice particle of similar curvature or aerosol particle) of radius $R$ (Fig. 1), and the geometry of its 2D analogue (Fig. 2), we see that the arc length of a small circle of radius $r_\mathrm{w}$, signifying the range of the intermolecular force and the corresponding sector of the van der Waals interaction sphere centered around a surface molecule, inside the line limiting the drop, is given by: $S_1 = 2r_\mathrm{w}$ arccos $r_\mathrm{w}/2R$. Considering that $R \gg r_\mathrm{w}$, the arccos term can be approximated by $1/2(\pi - R/r_\mathrm{w})$. Denoting the arc length of the sector of the ring of radius $r_\mathrm{w}$, which is outside the drop by $S_2$ then the difference between the arcs is $\Delta S = S_1 - S_2 = r_\mathrm{w}^2/R$. Introducing the ring asymmetry $A$, by relating $\Delta S$ to the circumference of the van der Waals interaction sphere we get: $A = \Delta S/2\pi r_\mathrm{w}$. On the basis of Eq. (2), the quantity $A$ has the physical meaning of a small correction of the

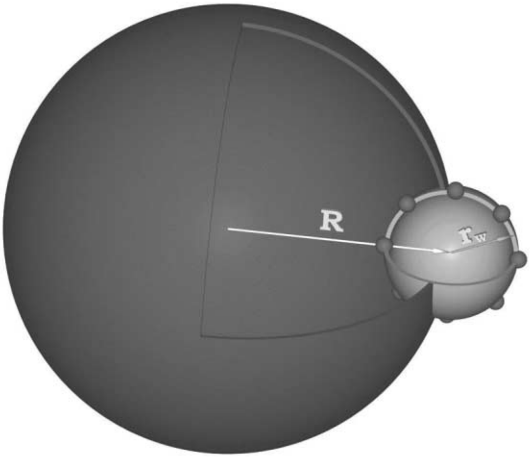

Fig. 1. A schematic representation of the 3D asymmetric spatial distribution of molecules affected by the interfacial force at curved, liquid coated surfaces. As can be seen, only three out of eight equidistantly distributed molecules (small spheres) are located inside the arc limiting the larger sphere.

coordination number $z_s$ accounting for the deviation of the shape of the curved surface from the planar situation:

$$E_s^o = \frac{1}{2} z_s (1 - A) W_{AA}. \tag{3}$$

Eq. (3) allows us to determine the sign of the difference of the interaction energies $\Delta E_s^o$ of two adjacent surface molecules belonging to two drops of radius $R_1$, $R_2$, or ring asymmetry $A_1$, $A_2$:

$$\Delta E_s^o = E_{s_1}^o - E_{s_2}^o. \tag{4}$$

Substituting the expression for $\Delta S$ into the expression defining the ring asymmetry $A$, we obtain the expression for $\Delta E_s^o$ on the basis of Eq. (3):

$$\Delta E_s^o = \frac{1}{4\pi} z_s r_w W_{AA} \left( \frac{1}{R_2} - \frac{1}{R_1} \right), \tag{5}$$

where $W_{AA}$ denotes the pair interaction energy, $r_w$ is the range of the attractive intermolecular force and $z_s$ is the coordination number of a surface molecule belonging to a plane liquid surface. This equation, denoting the free energy reduction by molecular interface crossing between curved liquid surfaces, has been rigorously derived for the 2D configuration and experimentally verified for nanoscale Perfluorocarbon drops in interfacial contact (Sommer et al., 1999). By noting that the pair interaction energy $W_{AA}$ is a negative energy, it is clear from the above equation that $\Delta E_s^o$ becomes negative for $R_1 > R_2$.

Following the definition of $\Delta E_s^o$, a negative $\Delta E_s^o$ is equivalent with $\Delta E_{s_2}^o > \Delta E_{s_1}^o$. Thus, a molecule at the surface of the smaller drop should be less stable, having a higher probability for fluctuations than the corresponding surface molecules from the larger

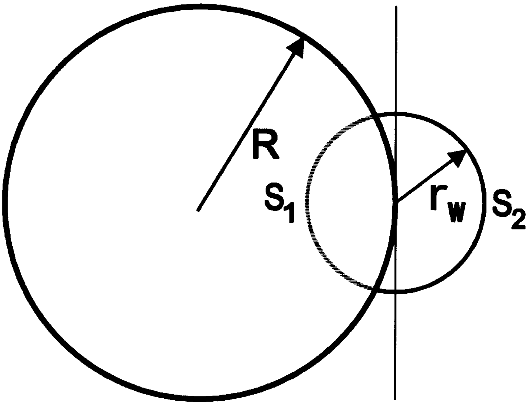

Fig. 2. Two-dimensional analogue of the 3D model shown in Fig. 1.

drop. For two droplets approaching each other to distances in the order of magnitude $r_w \ll R_1, R_2$, Eq. (5) predicts the transfer of molecules from the smaller to the larger drop. This is because the system tends to reduce its potential energy through its evolution towards the highest possible thermodynamic stability.

Interfacial transport observed for FERMIC has the same direction as characteristic for Ostwald ripening (Ostwald, 1900), a diffusion controlled transfer process working at large distances in the presence of spatial concentration gradients driving the molecules at intact boundaries from a smaller particle to the larger one.

Further evidence of the validity of intermolecular material transport induced by the FERMIC mechanism has been found recently during laboratory experiments using an atomic force microscope, AFM (Zahn, private communication). In these experiments, water molecules adhering to the surface of the nanoscopic silicone tip of the AFM were transferred to the crystalline plane surface below during scanning at a separation distance in the order of the tip diameter. This phenomenon could be explained by a mechanism similar to the transfer of water molecules between drops of different radii.

## 3. Application to cloud electrification

The extension of the FERMIC mechanism to the processes occurring between contacting ice particles coated with a water layer (molecular mask) seems clear. For distances in the range of the attractive intermolecular forces between water molecules (Israelachvili and Wennerström, 1996), the FERMIC model predicts the transfer of water molecules (and ions) in the energetically preferred direction from the liquid layer (or quasi-liquid layer) on the surface of the ice crystal to the lower curvature surface of the graupel. For interfacial separations between ice crystal molecules and molecules belonging to the graupel within the range of the attractive intermolecular forces, the asymmetrically bound water molecules (Perrin et al., 1998) and ions (Bianco et al., 1998; Cowin et al., 1999) at the surface of an ice crystal are expected to reach the surface of the graupel before the "massive" impact takes place. Thus, the binding of a water molecule on the surface of an ice crystal (air/water interface), in proximity of a graupel pellet, can be regarded as the sum of three competing contributions: the bonding to the nearest molecules in the liquid layer coating the crystal, a long range attraction between the air/water and the water/ice interfaces, and the attractive component induced by the powerful FERMIC mechanism, driving the asymmetrically bound molecules (and ions) towards the graupel surface. As the distance between the approaching two surfaces decreases, less bound mobile water molecules on the crystal surface are more and more attracted to the side of the graupel. At distances within the effective range $r_w$ of the intermolecular forces, the FERMIC mechanism becomes active forcing the transfer of interfacial water molecules from the ice crystal surface to the graupel. In order to specify the interaction energies, it would be beneficial to quantify the Hamaker constant for the liquid-like water film coating the crystal in the relevant temperature range. Prerequisites to calculate this quantity are the dielectric constant and the index of refraction for the supercooled liquid water film—parameters practically unknown in the published literature.

The dynamical mass transfer picture we regard here is quite different from the known vapour diffusion between two cloud particles. The direction of the first transfer phase, operating before interfacial contact and driven by the FERMIC process, is opposite to the transport route for the vapour transfer confirmed by current theory. During mass transfer induced by the FERMIC process, water molecules still belonging to the liquid-like layer at the highly curved ice crystal surface are driven towards the low curvature side of the graupel exclusively by intermolecular attraction. This transfer is active only before contact and is ineffective when the distance between the interacting surfaces is not within the specified near-field range. For distances corresponding to the range of the attractive intermolecular forces, i.e. the mean free path length of a water molecule crossing the low-density air space dividing the interface (Stecki and Toxvaerd, 1995), the transit time for water molecules or ions is expected to be considerably shorter than any value calculated from the bulk water diffusivity. Upon reaching a distance of interfacial separation of the order of 10 nm, the FERMIC field acts on the molecules and ions near the point of closest approach, causing them to be accelerated and to move from the surface of the ice crystal to that of the graupel.

Material from the high curvature surface film masking the dendritic ice crystal tip approaching a graupel at a velocity of about 1–5 m/s will land on the graupel before the impact. In the idealized model, simulating the interaction between a large graupel and a small ice crystal, the transfer of material before interfacial contact (vanishing at equal curvatures) increases according to Eq. (5) with increasing curvature asymmetry. Following the transfer (Kleyn, 1997) to the side of the graupel, the molecules and ions reduce their energy state reaching a higher thermodynamic stability. Our present preliminary model does not consider the effects of the LWC and of the cloud temperature, which may affect the crystal growth by diffusion (Baker et al., 1987) in the presence of moving boundaries and consequently influence the approach velocity of the particles. It is believed that this and possible local film deformations of the liquid-like layer coating the crystal have only a secondary effect on the strength of the FERMIC field.

Interestingly, the bulk/interface asymmetry in the intermolecular forces between the neutral water dipoles is enriched by a temporal asymmetry in the lifetimes of the two charged species that are coexisting in liquid water. Interpretations of nuclear magnetic resonance experiments, NMR, revealed a significant difference in the lifetime for prototropic ions in water (Halle and Karlström, 1983). The principal result of this study, probably of relevance in the understanding of the charge transfer mechanisms occurring during interactions of cloud particles in thunderclouds, is that the mean lifetime of an $H_3O^+$ ion is about half the mean lifetime of an $OH^-$ ion. This ratio was determined for a temperature of 28°C. Assuming no major structural differences between supercooled liquid water and water at 28°C, we suggest that charge transfer during interactions of ice crystals with graupel particles could be a result of such asymmetric molecular/ionic transfer across the contact interfaces. Since the ions are transferred together with water molecules from the smaller particle (ice crystal) to the larger one (graupel), those with longer lifetimes can manage to survive the transfer. Shorter-lived ions will be neutralized before moving across. In other words, $OH^-$ ions will preferentially be transferred from the ice crystal to the graupel, making the latter negatively charged and leaving the crystal positively charged, as observed in laboratory experiments. This principle result is also in agreement with observations in nature in which most frequently the lower (containing the larger particles) and upper (containing the smaller ice crystals) cloud charge centres are negatively and positively charged, respectively. Due to the range of the intermolecular field across the interface ($h \sim 10$ nm), any ion created prior to contact at the higher curvature surface will be attracted towards the lower curvature side. On account of the mean lifetime of an $OH^-$ ion of about $\tau \sim 10^{-12}$ s (Halle and Karlström, 1983), formed at the surface of the liquid-like layer on the ice crystal at an interfacial separation from the graupel of the order of $h$, the minimum velocity for such an ion required to reach the graupel is about $10^4$ m/s. This velocity corresponds to a kinetic energy of the order of 400 kT, where kT is the thermal energy at $-15$°C and the thermal energy induced fluctuation of a surface molecule at the liquid-like layer is assumed to be proportional to kT. For an interfacial separation of the order of 10 Å, the ion requires a velocity in the order of $10^3$ m/s to cross the interface, corresponding to a kinetic energy of about 4 kT. Thus, for a low-density interfacial gap within the range of the intermolecular field (10 Å–10 nm), the energy necessary to remove ions from the ice crystal side could be provided by the FERMIC energy difference. The distance, which a free ion formed at an ice crystal surface may travel before neutralisation, is equal to about 5 Å, corresponding to the diameter of two $H_20$ molecules. Under the influence of an energy difference provided by the FERMIC process, the ions will have no difficulty in moving from the ice crystal to the graupel particle. Due to the motion of the ice particles in the thundercloud and the limited time window for the transfer processes, the charge resulting from crossing and deposition of ions in the first transfer phase (before contact) will probably not be neutralized on contact.

As a first approximation, the charge transferred via ice particle contacts observed in laboratory experiments is around 1 fC. In order to estimate the order of magnitude of the charge transferred between two asymmetrically curved surfaces before contact, we start from an ice crystal dendrite tip sector in the order of 0.1 μ and a mean distance between two surface charge centers of 5 Å. Let us further assume a distance between the ice crystal dendrite tip and the graupel surface in the 10 Å–10 nm range, ensuring that molecules and ions will be transferred via FERMIC and that the charge transferred to the graupel stems from the uppermost liquid-like layer on the dendrite tip of the ice crystal. Since transfer time and mean lifetime of the ions are of the same order, we assume that every second surface molecule has a chance to be transferred as a negative ion, half of which is being neutralized upon transfer. The resulting number $(4 \times 10^4)$ of negative ions transferred from the selected dendrite tip sector to the graupel corresponds to about 6 fC—the order of the charge that is likely to be separated on contact. Compared to the bulk, the natural ice/ice interfaces are highly contaminated with ionic and neutral molecules, either coming from the gas phase or exuded from the bulk ice, inducing high diffusional mobilities and conductivities. Thus, the charge transferred in the cloud could increase sensitively compared to the estimated value.

The FERMIC model accounts for the observation that charge transfer resulting from ice crystal/ice crystal collisions and bounce will not be large because of the similar geometry of the particles. To complete the picture, a simple argument why charge build-up is not observed during drop/drop collisions should be added. Measurements show that the collision of small cloud drops more often result in collection than in bounce. This implies that charge build-up would be small due to such collisions. Bounce is more probable during collisions of large drops (e.g. Low and List, 1982), but their concentrations are low and would probably not contribute much to the charge separation process. Therefore, although the paradigm we propose here is expected to be valid in principle during collisions and bounce of water drops too, the probability for transferring charge in clouds by the drop collisions is expected to be small.

In its present form, our primitive model does not cover the observed charge-reversal at higher thundercloud temperatures that was reproduced in laboratory experiments by Takahashi (1978) and discussed by Williams (1989) and Williams et al. (1991). Consequently, additional mechanisms accounting for the relative ice/graupel temperature difference, potentially driving the mobile ions in the opposite direction have to be included. While FERMIC could be the mechanism dominating interfacial mass and charge transfer processes at lower thundercloud temperatures, it is reasonable to assume that other mechanisms (e.g. Jayaratne et al., 1983; Saunders et al., 1991; Saunders, 1993; Brooks et al., 1997; Avila et al., 1998; Saunders and Peck, 1998; Avila et al., 1999; Saunders et al., 1999; Pereyra et al., 2000) could control transfer processes at higher cloud temperatures. With increasing temperature, local curvature differences between ice crystal and graupel pellet are likely to diminish, principally due to melting processes at the ice crystal surface. From Eq. (5), it becomes evident that local melting will be accompanied by a decreasing geometrical asymmetry between the cloud particles, with a corresponding sensitive decrease in the driving force stemming from the FERMIC energy difference. In addition, temperature increase is generally associated with a decrease in surface tension. The smaller the surface tension, the lower the potential energy and so the energy barrier that has to be surpassed by the mobile molecules/ions on either surface. Thus, not only the thickness of the liquid-like layer on the ice particles is expected to increase with increasing (cloud) temperature (Döppenschmidt and Butt, 2000), but also the vapour pressure $p$ in the interfacial gap: $p(-1°\text{C}) \sim 3p(-15°\text{C})$. This could mean that more water molecules fill the interfacial gap at higher cloud temperatures, potentially neutralizing some of the ions that cross the gap. In addition, thicker water layers on the ice particles could result in less bounce between the colliding hydrometeors, compared to lower temperatures. Less bounce obviously means lower charging efficiency.

In an advanced unified thundercloud electrification model, including charge transfer occurring at different temporal phases of the collision, the balanced interplay between various relevant counteracting forces, temperature gradients at the contact interface and LWC sensitive interfacial effects could presumably not be satisfactorily solved without computer simulations. In summary, it is worth noting that the majority of the data presently available which are related to molecular transfer processes—even for simplest liquid/liquid interfaces—stem from computer simulations (Benjamin, 1992; Hynes, 1999; Marx et al., 1999), and not from experimental observations (Richmond, 1997). Thus far, these calculations are extremely successful to account for charge and mass transfer across liquid interfaces under stationary conditions. However, using suitable potentials (Israelachvili and Berman, 1995; Heilweil, 1999) for the interfacial space and adequate Lennard–Jones terms for the intermolecular interactions, it should be possible to shed more light on the charging mechanisms in thunderclouds—a significant challenge to scientists trying to unravel the complexity of the electrical processes in the atmosphere.

## 4. Conclusions

The present paper discusses a new molecular mechanism for charge transfer in clouds. The mechanism, Free Energy Reduction by Molecular Interface Crossing (FERMIC), is based on theoretical calculations and laboratory experiments that show that molecules and ions can be transferred between two approaching surfaces (water drops or ice crystals with water layers on them), when the distance of separation is in the range of the mean free path. The molecules and ions are transferred from the smaller cloud particle to the larger one and charge can be separated, provided the contact time is shorter than the neutralization of the charge. Since $OH^-$ ions have longer mean lifetimes than $H_3O^+$ ions, negative charges will be transferred preferentially from the liquid-like layer on the ice crystals to the graupel particles, leaving the ice crystals positively charged and the graupel negative, as reported for a majority of thunderclouds at $-15$°C. This is the picture resulting from the simplest model. Further clarification is needed from the experimental side and could be expected from nanoscale imaging of the ice particle surfaces. AFM inspections of natural and synthetic ice particle topography, examined at different temperatures, may help to get access to the geometrical situation on natural ice particles.

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