

Charge transfer process during ice-ice collision within a thundercloud

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Formation of a quasi solid layer (QSL) over the ice substrate at low temperatures due to riming is assumed. A mechanism of charge transfer in ice-ice collision is proposed based on this QSL formalism. The amount (Δq) and sign of charge transfer are supposed to depend on the relative depth of QSL of two colliding bodies. Variations of Δq on various parameters are studied. The estimated order of magnitude of Δq is found to lie between 10^{-15} C and 10^{-12} C. The QSL formalism is found to be applicable only below a transition temperature specified by the local conditions and sizes of the particles.

1 Introduction

The worldwide location of thundercloud charge centres at a temperature below the freezing level temperature implies the importance of ice phase in the charging process. Substantial charge is found to be transferred during interaction of ice crystals with riming graupel pellets in presence of supercooled water droplets¹.

Several laboratory simulations have been carried out to estimate the magnitude and sign of the charge transfer during ice-ice collision. However, the charge transfer was found to depend on local conditions. Takahashi² noted the dependence of sign of charge transfer on the temperature as well as liquid water content (LWC) of the cloud. On the basis of laboratory observations, Jayaratne *et al.*³ could identify the existence of a reversal temperature (T_r). At temperature above T_r the graupel is charged positively, whereas below T_r the graupel is charged negatively. This T_r was found to shift to lower temperatures for higher LWC and vice-versa. Based on this reversal temperature concept, the tripole structure of thunderstorm⁴ can be satisfactorily explained. Keith and Saunders⁵ extended their work to larger ice crystals and observed a dependence of charge transfer also on the size and fall speed of the rime.

Saunders *et al.*⁶ noted that the effective liquid water content (EW), which is the portion of the droplet spectrum actually captured by the rime, is a better parameter than the LWC. Based on their experimental results they developed a series of empirical relations for the charge transfer in terms of temperature, EW, fall speed, and size of the crystals.

Incidentally, all these quantifications are mainly

empirical. Debate is still going on about the exact physical mechanism that underlies the charge transfer process. A possible mechanism for charge transfer during the interaction of ice crystal and graupel is outlined by Baker and Dash⁷ who introduced the concept of surface melted liquid like layer over the ice surface. They termed the surface melted liquid like layer as quasi liquid layer (QLL). This QLL was supposed to contain dissociated hydroxyl and hydronium ions (when no other contaminants are present). During contact, the QLL fluid flows from a thicker to a thinner layer and thereby losing net charge from one body to the other. The depth of this QLL was assumed to depend on the local conditions and sizes of the particles. Their estimate of charge transfer was found to have overall consistency with laboratory observations.

In Baker and Dash⁷ model, the ice particles were assumed to grow by diffusion of water vapour and the role of suspended supercooled droplets in the atmosphere was not considered. But, laboratory experiments show that the presence of these supercooled droplets is necessary for appreciable charging. Another disadvantage of their model is that at temperatures much below the melting temperature, surface melting cannot occur. Further, there is experimental evidence⁸ that a liquid like layer on ice surface no more exists below -4°C . Thus, at lower temperatures, some different mechanism other than this QLL hypothesis should be effective.

In this paper, a probable mechanism of charge transfer at lower temperatures is proposed. Here, the role of the supercooled liquid droplets is considered explicitly. Formation of a disordered sur-

face layer, viz. quasi solid layer (QSL) due to rimming is considered⁹. The equilibrium depth of this layer is calculated from the free energy consideration. In the proposed model the transfer of this QSL during collision actually accounts for the charge transfer. The magnitude of charge transfer in this process is estimated and its dependence on various environmental parameters is studied.

2 Charge carriers in QSL

Keith and Saunders⁵ proposed that a possible charge transfer mechanism may involve dislocations and associated charges on the crystal and graupel surface. In the present work, the charge transfer problem is considered from this point of view. Here, the charge carriers are supposed to be the defects formed during rimming. Droplets freeze faster with decrease in temperature¹⁰. This rapid freezing of droplets on the ice substrate gives rise to the formation of defects in the lattice structure. The number density of defects formed depends on the growth rate and freezing characteristics⁵. In the present work the defects are considered to be formed mainly by the mechanical stress due to the impact and also due to the rapid freezing at low temperature.

2.1 Nature of defects

In the temperature region of our interest generally hexagonal ice crystals are formed¹¹. In general, the oxygens are crystallographically arranged and the protons are randomly distributed¹² in the lattice. This distribution is consistent with the two rules of Bernal and Fowler. Any breach of these rules gives rise to the formation of structural defects in ice.

The equilibrium concentration of ionic defects (3×10^{-12} mol of def./mol of ice) is generally very low compared to that of orientational defects (2×10^{-7} mol of def./mol of ice)¹³. The orientational defects as proposed by Bjerrum are of two types, viz. *L* and *D* (Ref. 12). The presence of two protons over an O-O bond is termed as a *D*-defect, whereas an O-O bond without any proton means a *L*-defect. Dunitz¹⁴ has shown that the electrostatic repulsion between two collinear protons in a *D*-defect (following Bjerrum's geometry) makes its formation rather improbable. Instead another type of defect called *X*-defect, which appears in pair with *L*-defect, was proposed by him. In order to maintain the charge stoichiometry, these *X*-defects are positively charged defects. The existence of this *L-X* pair is being considered to be very common during the growth of crystals.

Again, the mechanical stress due to the impact of droplets on the ice substrate may activate the self-diffusion mechanism. Self-diffusion is believed to occur due to the migration of entire water molecule within the lattice, creating vacancies at certain lattice positions¹⁵. Onsager and Runnels¹⁶ proposed that during self-diffusion, the water molecules traverse several lattice distance through the interstitial space and then occupies a normal lattice position. They have also shown that the migration of diffusing molecules is faster by one order of magnitude to migration of the orientational defects.

Now, it may so happen that during its journey through the interstitial space the diffusing molecule encounters a *L*-defect. The lone pair electrons in the *L* will show an affinity to form hydrogen bond with this interstitial molecule. Thus an interstitial associate¹⁷ (*LI* associate) will form, thereby immobilizing the diffusing molecule and leaving a vacancy and a relatively inactive positive *X*-defect¹⁴ in the lattice. The vacancies so formed in the rimming process will reduce the density of the rimmed ice appreciably¹⁸. A further consequence of our assumptions in the model is that there is no free negative ions available in the lattice to crowd around the positive charge so as to neutralize it. Thus, remaining *X*-defects will be the charge carriers in the mass transfer process during collision. The number density of the available positive charge carriers will be equal to the number density of *LI* associates formed.

2.2 Number density of defects

If the rimming process continues, successive layers with positive defects will be deposited. The defects are more prone to form over a disturbed surface rather than on a regular one. As the process continues, the number of defects formed in successive layers will increase, thereby creating a concentration gradient across the depth of the rimmed layer. An exponential relation may be considered in the form

$$n(z) = n_s \exp(-z/h_m) \quad \dots (1)$$

where the penetration depth is measured from the top of the surface, n_s is the concentration of defects at the topmost layer, $n(z)$ is that at a depth z , and the scaling factor h_m is the maximum possible depth of rimmed mass under a specific condition over an ice particle. The scaling factor h_m is estimated from a relative growth rate calculation⁹.

Again, since the defects are considered to form due to rimming, the number of defects formed is a

function of rate of accretion. For maximum possible rate of accretion α_m , the number of defects formed will be maximum. Here α_m is given by

$$\alpha_m = w_m u_m r_m^2$$

where

r_m Maximum size of particles

w_m Maximum possible water content of cloud

u_m Maximum possible velocity of particles

We here assume that no defect will form without accretion. Keeping this view in mind a relation between accretion rate and n_s can be written as

$$n_s = n_m \{1 - \beta \exp(-\alpha/\alpha_m)\} \quad \dots (2)$$

where

$$\alpha = w u r^2$$

$$\beta = 1 - \alpha/\alpha_m$$

The maximum number of defects is taken as the equilibrium concentration of defects¹⁷ and the distribution is given by

$$n_m = N_0 \exp(-Q_{LI}/kT) \quad \dots (3)$$

where

N_0 Number of molecules per unit volume

Q_{LI} Formation energy of LI associates

k Boltzmann constant

T Absolute temperature

3 Charge transfer process

It is assumed that during collision, the soft QSL part containing positive defects is shared by the two colliding bodies across the contact interface resulting in a transfer of charge after separation. During riming the conductivity of ice particles increases¹⁹. Thus there is sufficient time for the transferred charge to be redistributed over the colliding rimed particle surface. As a result, large field cannot be developed during separation to initiate return flow of charge by corona as proposed by Keith and Saunders¹⁰.

The particle with thicker QSL will lose the mass and the one with thinner QSL will gain the mass. The former particle will thereby gain a negative charge and the latter will gain a positive charge. The sign of charge transfer will then be a function of relative growth rate which is consistent

with the proposition of Baker *et al.*¹⁰ The magnitude of charge transfer will depend on the depth of QSL containing defects and the area of the contact interface. The equilibrium depth of QSL containing defects is estimated from a free energy consideration.

4 Calculation of h_{QSL} and Δq

4.1 Assumptions

Before going further the assumptions are being clearly stated:

(i) All the particles are considered to be spherical.

(ii) Collection efficiency of droplets by the ice particles is cent per cent.

(iii) All through the cloud column distribution of liquid water is uniform.

(iv) Due to the formation of vacancies and interstitials there will be some redistribution of layer charge which will give rise to some electrostatic interaction. Here, only the dipole interaction is considered and the higher order terms are neglected.

(v) Since the elastic constants of rime are not available in the literature, it is further assumed that the estimated elastic moduli of crystalline ice equally apply to rimed ice.

4.2 Determination of h_{QSL} by minimization of free energy term

The probability (P) of formation of $n LI$ associates is the product of the probability (W) of formation of n interstitials (I) and probability of association (A) of I with L defects. Therefore

$$W = [N!/(N-n)! n!] [N_i!/(N_i-n)! n!] \quad \dots (4a)$$

where N and N_i are the available numbers of lattice and interstitial sites respectively. Since N_i is almost of the order of N one can write

$$W = [N!/(N-n)! n!]^2 \quad \dots (4b)$$

Since an I will either associate with one of the N_L number of L defects or remain free, A is given by

$$A = 1/2N_L \quad \dots (4c)$$

Therefore

$$P = WA = [N!/(N-n)! n!]^2 [1/2N_L] \quad \dots (4d)$$

At a depth z , the change in free energy due to the formation of defects is given by

$$(\Delta F)_z = n(z) Q_{LI} - kT_{av} \ln P \quad \dots (5a)$$

where

$$T_{av} = (T + T_s)/2 \quad \dots (5b)$$

Here T is the absolute temperature, and T_s is the surface temperature of the ice particle which is calculated following the heat balance equation given by Macklin and Payne²¹. $n(z)$ is given by Eq. (1).

Following Eq. (4d) and assuming $N \gg n$, one can write

$$\ln P = 2n(z) \ln N - 2n(z) \ln [n(z)] - \ln 2 - \ln N_L \quad \dots (6)$$

However, N_L is given by

$$N_L = N \exp(-Q_L/kT_{av}) \quad \dots (7)$$

where Q_L is the formation energy of L defects. Combining Eqs (3a), (5), (6) and (1), one obtains

$$\begin{aligned} (\Delta F_f)_z = \{ n_s \exp(-z/h_m) Q_L - 2n_s kT_{av} \exp(-z/h_m) \\ \times [\ln N - \ln n_s + z/h_m] - kT_{av} (\ln 2 + \ln N) \\ + Q_L \} \quad \dots (8a) \end{aligned}$$

As already mentioned, there will be some electrostatic interaction which is to be included in the free energy calculations. The dipole interaction is only considered here. Dipole moment due to a single defect formation is $\mu = e_d x/\sqrt{2}$, where x is the distance between the two consecutive layers, and e_d is the charge on the defect and is given by $0.2 e$, e being the electronic charge. The net dipole moment is given by $M = n(z)\mu$, and the associated electric field is given by $E = 4\pi M/\kappa$, where κ is the dielectric constant of ice. κ is calculated following Eisenberg and Kauzmann²². The interaction energy is then given by $\Delta U = ME$. Thus, the free energy change at a depth z is given by

$$(\Delta F)_z = (\Delta F_f)_z + \Delta U \quad \dots (8b)$$

Therefore, net free energy change due to the whole layer of depth h is

$$\begin{aligned} \Delta F = \int_0^h (\Delta F)_z dz \\ = Ah + B[1 - \exp(-h/h_m)] + Ch \exp(-h/h_m) \\ + D[1 - \exp(-2h/h_m)] \quad \dots (9) \end{aligned}$$

where

$$\begin{aligned} A = Q_L - kT_{av} \ln 2 - kT_{av} \ln N \\ B = h_m \{ n_s Q_L - 2n_s kT_{av} \ln N + 2n_s kT_{av} \ln n_s \\ - 2n_s kT_{av} \} \end{aligned}$$

$$C = 2n_s kT_{av}$$

$$D = 4\pi e_d^2 x^2 n_s^2 h_m / \kappa$$

The equilibrium depth of QSL (h_{QSL}) may be obtained minimizing ΔF with respect to h .

4.3 Estimation of Δq

The net charge transfer during collision of two particles is

$$\Delta q = \bar{n} e_d (h_{QSL2} - h_{QSL1}) \pi a^2 \quad \dots (10)$$

where

$$\bar{n} = (\bar{n}_1 + \bar{n}_2)/2$$

$\bar{n}_i$ Average number density of defects for particle i

h_{QSLi} h_{QSL} for particle i ($i = 1, 2$)

a Radius for the contact area

The radius of the contact area (a) is calculated following Hertz theory of impact²³ and is given by

$$\begin{aligned} a = (v/V_c)^{0.4} \{ (r_1 r_2)^4 / (r_1 + r_2)(r_1^3 + r_2^3) \}^{0.2} \\ \times \{ 5\pi(1 - \sigma)^2 / 2(1 - 2\sigma) \}^{0.2} \} \end{aligned}$$

where r_i is the radius of the particle i ($i = 1, 2$), σ the Poisson's ratio, v the velocity of impact, and V_c is given by $V_c = \lambda + 2\mu_{el}/\rho$, when λ and μ_{el} are Lamé coefficients and ρ is the density. The elastic constants at different temperatures are calculated following Danti²⁴. These are used to calculate σ , λ and μ_{el} (Ref. 25).

5 Results and discussion

Table 1 represents the values of h_{QSL} for particles of different sizes, fall speeds (u) and w . The cloud temperature is taken to be -10.0°C . As the sign of charge transfer is determined by the thick-

Table 1— h_{QSL} values for different values of r , u and w at $T = -10.0^\circ\text{C}$

w gm ⁻³	u ms ⁻¹	h_{QSL} (μm)			
		$r = 100 \mu\text{m}$	$r = 250 \mu\text{m}$	$r = 500 \mu\text{m}$	$r = 5000 \mu\text{m}$
1.0	3.0	4.42	4.08	8.81	2.77
	5.0	9.92	9.13	8.49	6.05
	7.0	14.20	13.10	12.10	8.51
	9.0	18.50	16.90	15.70	10.90
3.0	3.0	14.10	12.90	11.90	8.30
	5.0	32.70	29.80	27.50	18.70
	7.0	44.00	44.00	36.80	24.60
	9.0	56.00	50.80	46.60	30.80

Table 2—Values of Δq for graupel of radius 5.0 mm at $T = -10.0^\circ\text{C}$ and $w = 1.0 \text{ gm}^{-3}$ for various sizes of interacting ice crystals for different values of u and u_d

r μm	u_d ms^{-1}	$\Delta q (\text{fC})$		
		$u = 5.0 \text{ ms}^{-1}$	$u = 7.0 \text{ ms}^{-1}$	$u = 9.0 \text{ ms}^{-1}$
50	3.0	0.021	0.142	0.408
	5.0	0.000	-0.049	0.022
	7.0	-0.183	0.000	-0.151
	9.0	-0.531	-0.347	0.000
100	3.0	0.070	0.425	1.190
	5.0	0.000	-0.091	0.141
	7.0	-1.404	0.000	-0.290
	9.0	-1.140	-1.730	0.000
250	3.0	0.348	1.890	5.140
	5.0	0.000	-0.157	0.995
	7.0	-1.320	0.000	-0.728
	9.0	-3.610	-2.230	0.000

ness of QSL on individual particles, this table indicates that at a fixed environmental condition the sign and magnitude of the charge transfer may be more sensitive to the respective speeds [fall speeds in case of large particles (u) and draft speed (u_d) in case of small particles] of the individual particles than to the relative impact velocity. This fact can easily be realized from Table 2.

Table 2 shows that the magnitude of Δq increases with increasing particle size. This is quite obvious since the contact area in that case increases. In our calculations the effective water accreted is directly proportional to the respective fall speed (u) of the particle. Since h_{QSL} is actually dependent on the effective liquid water collected, the associated charge transfer also shows more dependence on the respective fall speed of the particles rather than on the relative velocity of impact.

However, if the interacting crystal speed (u_d) is kept constant, it is found that the charge transfer is found to increase with the increase in impact velocity (Table 3). Δq is also found to increase with increasing w .

It is to be noted here that the calculated order of magnitude of Δq (10^{-15} C to 10^{-12} C) agrees approximately with the experimental observations. Appreciable amount of charge is found to be transferred at temperature down to -35.0°C (Fig. 1). At low liquid water content the riming is not appreciable and thus the charge transfer is also low.

Calculations based on growth rate show that in general above -10.0°C temperature the QSL concept is not suitable. Above that temperature there is probability of surface melting and the conse-

Table 3—Values of Δq for graupel of radius 5.0 mm at $T = -10.0^\circ\text{C}$ and $u = 3.0 \text{ ms}^{-1}$ for various sizes of interacting ice crystals for different values of w and v

w gm^{-3}	v ms^{-1}	$\Delta q (\text{fC})$		
		$r = 50 \mu\text{m}$	$r = 100 \mu\text{m}$	$r = 250 \mu\text{m}$
1.0	2.0	0.021	0.070	0.348
	4.0	0.142	0.425	1.890
	6.0	0.408	1.190	5.140
3.0	2.0	0.222	0.659	3.130
	4.0	1.350	3.590	15.000
	6.0	3.850	9.940	40.100
5.0	2.0	2.170	4.990	19.100
	4.0	11.800	24.200	82.400
	6.0	34.600	68.400	222.000

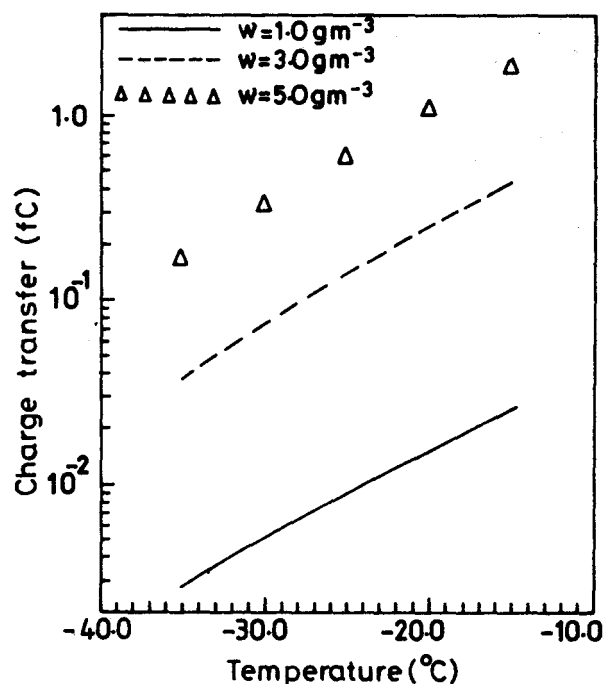


Fig. 1—Temperature dependence of charge transfer during collision of a graupel of radius 1.0 mm, falling with a velocity of 6.0 ms^{-1} with a crystal of radius $50 \mu\text{m}$ moving with the updraft with a speed of 3.0 ms^{-1} .

quent formation of liquid like layer modifying the character of the surface appreciably. Depending on the effective liquid water content this transition temperature might shift even to -15.0°C . Thus the proposed charge transfer mechanism will be applicable only to the temperature regime below a transition temperature dependent upon specific atmospheric conditions.

It should be noted that in the case of liquid like layer formalism⁷ the sign of the charge transfer is

of opposite polarity to that considered in the present work. So the transition temperature noted may carry some implication to the well known charge sign reversal temperature³.

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