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Collisional charging in ice and charge separation in thunderstorms

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ABSTRACT

LABORATORY AND FIELD STUDIES INDICATE THAT THUNDERSTORM ELECTRIFICATION CAN BE ATTRIBUTED TO REBOUNDING COLLISIONS BETWEEN SMALL ICE PARTICLES AND HAIL. THE FUNDAMENTAL MECHANISM OF CHARGE SEPARATION HAS DEFIED EXPLANATION FOR MANY YEARS, BUT RECENT EXPERIMENTS OFFER NEW CLUES, LEADING TO A QUANTITATIVE THEORY OF THE MICROPHYSICAL PROCESS. THE THEORY EXPLAINS THE SYSTEMATIC DEPENDENCE OF CHARGE AND MASS TRANSFER ON TEMPERATURE, COLLISION SPEEDS AND GROWTH RATES THAT HAVE BEEN OBSERVED IN LABORATORY STUDIES. APPLIED TO STORM DYNAMICS, THE THEORY DESCRIBES HOW THE UPPER PORTIONS OF THUNDERCLOUDS BECOME POSITIVELY CHARGED, WHILE THE NEGATIVE CHARGE RESIDES ON HAIL IN THE LOWER REGIONS.

INTRODUCTION

The study of thunderstorms has revealed considerable details of their electrical nature (1). The anatomy of a common thundercloud shows a principally dipolar charge distribution, having a positively charged upper portion and a negative lower region. Negative lightning strokes originate in this layer, where active ice is generated. Many simulation experiments indicate that collisions between small ice particles and hail can account for the observed electric fields. The principal features collected from storm and laboratory studies are illustrated in Figs.1a and 1b.

Many laboratory studies have explored the systematic features of collisional charging, and several mechanisms have been proposed to interpret the findings. A detailed review (2) has shown that the proposed models have had limited success in modeling the complex dependence on environmental conditions. However, a recent experiment (3), hereafter referred to as MD, has provided additional clues, leading to a detailed theory of the molecular process (4).

The theory describes collisional charging as a three-part sequence; before collision, contact, and separation. Drawing on the field of surface physics and chemistry, the theory treats each stage in terms of the molecular dynamics of the ice interfaces. The detailed model explains the systematic trends with growth rate, temperature and collision speeds. In the following sections we outline the theory and compare its predictions with laboratory results and storm measurements.

THEORY

BEFORE COLLISION: PARTICLE GROWTH

The electrical nature of the ice particles stems from their growth history before collision. Well ordered crystals can be grown from vapor if the growth rate is slow enough for the deposited particles to diffuse on smooth surface facets to well separated attachment sites at steps, kinks and ledges. However, if the deposition rate is too fast the particles will nucleate into clusters at shorter distances, a process known as kinetic roughening (5). The scale of roughness is set by a competition between the deposition rate and surface diffusion.

The average time τ between arrivals of a vapor atom at a surface site is related to the site area *a* and deposition rate F by $\tau = (aF)^{-1}$. If the surface diffusion coefficient is D_s , then during τ a freshly deposited molecule travels an average distance $\lambda = (2\, D_s/\tau)^{1/2}$ (6) before being buried by another molecule from the vapor. Therefore the average distance between clusters is

$$(2\, D_s/aF)^{1/2}. \qquad (1)$$

In the case of ice, disordered surfaces contain ionic defects at corners and edges. The ionization leaves (OH-) ions bound to the site by their remaining hydrogen bonds, while the mobile positive ions diffuse deeper into the ice, thus creating a charged double layer and a negative surface potential. The surface charge density is proportional to the surface defect density: with Eq.(1), we have

$$\sigma/\lambda^2 = \xi\, e\, aF/2\, D_s \qquad (2)$$

where e is the elementary charge. The empirical parameter ξ is the surface structural disorder factor, which can depend on growth conditions and history. Comparison with experimental results yields the empirical value $\xi = 4$ for the conditions of the MD study.

The charged double layer in the ice is similar to the phenomenon of an ionized surface in liquid water (7,8). The diffusion of ions into the bulk is driven by the entropy available in the larger number of sites in the interior, and is opposed by the electrostatic field between the two distributions. This competition produces a concentration profile characterized by a Debye length κ^{-1}

$$\kappa^{-1} = (\varepsilon\varepsilon_o k_B T/e^2 n)^{1/2}, \qquad (3)$$

where k_B is Boltzmann's constant, ε is the dielectric constant of the medium, ε_o is the vacuum permittivity, and n is the background density of counterions. The results of a detailed calculation of the distribution and its dependence on growth rate are shown in Figs.2a and 2b.

DURING CONTACT:COLLISIONAL MELTING-

The impact between two ice particles causes temporary melting of a thin interfacial region (4). An estimate of the melt thickness is given by a simple expression,

$$d_m = \Delta E/Aqt, \qquad (4)$$

where ΔE is the inelastic energy loss, A is the area of contact, q is the latent heat of fusion, and t is the reduced temperature relative to the melting point T_o,

$$t = (T_o - T)/\, T_o. \qquad (5)$$

Equation 4 is derived by an extension of the theory of surface melting, which describes an equilibrium phenomenon common to most solids (9). In surface melting, the interface between a solid and vapor melts in a molecularly thin layer at a temperature below the bulk transition temperature, and then thickens as the temperature rises. In some materials surface melting begins as low as $0.8T_o$ but in ice without impurities the start is only a few degrees below 0^oC. Variants of surface melting occur at interfaces between a solid and a foreign substance, and at grain boundaries between crystals of the same

substance. Surface melting is enhanced by damage and other forms of disorder, which can cause melting at lower temperature or increase the thickness of the melted layer. The theory predicts that damage incurred in ice-ice collisions can cause melting of many molecular layers at temperatures well below 0^oC. It provides a quantitative explanation for the measured mass transfer in ice-ice collisions in the MD study.

Collisional melting between two ice particles destroys the site bonds of the surface (OH-) ions, and allows them to diffuse in the liquid layer. The brief time of contact, about 0.1 millisecond in the MD study, is sufficient for complete mixing of the (OH-) ions in the liquid layer, but too short for most of the positive ions to escape from their deeper sites in the solid. The contact is also too brief to allow much refreezing of the collisionally melted liquid.

WITHDRAWAL : DIVISION OF CHARGE AND MASS-

As two particles separate they take roughly equal shares of the collisionally melted liquid. The precise division will depend on details such as local shape and surface roughness, so that the sharing can only be estimated as a statistical average. Accordingly, in an average encounter the particles share the liquid mass, together with its dissolved ions, so that the one that had been growing more rapidly loses a greater charge. In typical cases the colliding particles have quite different histories; the more rapidly growing particle having been colder. Their temperature difference causes an asymmetry during the collision. Assuming equal sharing of the impact energy loss, Eq.4 predicts that the colder particle suffers less collisional melting. Consequently, when the melted liquid is shared, the colder particle comes away with some of the warmer particle's mass.

APPLICATIONS

In this section we test the theory against a checklist of systematic results of laboratory studies and storm observations.

COMPARISONS WITH LABORATORY RESULTS

Charge polarity – One of the strongest clues that helped the development of the theory was a conclusion drawn from a collisional charging study, that the particle which had been undergoing more rapid vapor growth is charged positively (10). The finding has been confirmed in several subsequent experiments (11). Additional details from the MD study show that the charge transfer is proportional to the rate of growth at low rates, but increases more slowly and declines at increasingly high growth rate. These results are shown in Fig.3, together with a theoretical curve. The agreement is obtained by fitting a single adjustable constant, the surface structural order parameter ξ. We note that the MD data are not simply fit by a scaling adjustment of ξ. We also point out that ξ is not simply a scaling parameter, for it appears in the theory in an essentially transcendental way.

The fall off at high rate is explained by the theory. At increasingly high growth rates the greater electric field between the positive and negative charge distributions limits the depth of the positive distribution. Due to its shallower depth, a larger fraction of the positive charge is liberated into the melt layer, thereby neutralizing more negative charge.

Identification of the mobile charge – Positive charging of the faster grown particle can be caused by acquisition of positive ions or by a loss of negative ions. Experiments on collisional charge transfer between ice particles and between ice and other materials has indicated that the transferred charge is negative (12). The theory agrees, in that the positive charging is due to the loss of (OH-) ions.

Linkage of surface charge and disorder- Experiments have shown that ice surfaces that have been damaged by abrasion or subjected to rapid growth by riming have large negative surface potentials (13). The theory is in agreement: ionization is caused by kinetic roughening, and the separation of positive and negative charges results in a negative surface polarity .

Mass transfer – The MD experiment shows that mass transfer accompanies charge transfer, the colder particle gaining an amount of mass roughly proportional to the temperature difference. The theory is in quantitative agreement with the measured mass transfer.

Fluidity of transferred mass- The MD study indicated that the mass exchanged during collisions was fluid like rather than solid fragments, even at temperatures as low as –16°C. This was an important clue in the development of the theory, for it could not be explained in terms of equilibrium surface melted fluid. It eventually led to an extension of the theory to non equilibrium effects and collisional melting.

COMPARISONS WITH STORM MEASUREMENTS

Thunderstorm dynamics indicate that small growing particles rising in updrafts are positively charged, and that the negative charges are on falling graupel particles. The theory of collisional charging explains this polarity as due to faster vapor growth on the small particles. At present, this interpretation is only a presumption, for the two particles grow by complicated mechanisms. The small particles probably experience rapid vapor growth immediately after freezing is nucleated, with a sudden burst of vapor when its temperature rises and then falls. Freezing occurs during their ascent; vapor growth then results from recapture of some of the particle's vapor and condensation from supercooled liquid droplets. The falling graupel, on the other hand, can grow by riming. The impact of supercooled liquid droplets nucleates freezing, which causes a sudden rise of temperature to the ice point, and a quick burst of vapor. A detailed calculation (10) shows that an appreciable fraction of the vapor recondenses on the graupel near the impact site. Depending on the surface temperature, the condensation can be very rapid. The collision region is therefore a mixture, where small particles and graupel can have either polarity, depending on their immediate histories and the details of their collsions. Such a mixture has been concluded from storm measurements (1), and is illustrated in Fig.1b.

The average charge density measured in typical storms is a result of quantitative difference between the populations. Judging the dominant polarity of collisional charging from the fact that most storms produce negative lightning from their lower regions, Fig.1b shows a representative case. In less common storms, positive lightning originates in lower layers of the cloud. This can be due to differences in cloud conditions, although the basic physics of collisional charging remain the same.

The theory requires temporary melting during contact in order to allow appreciable charge transfer. Model calculations confirm that collisional melting occurs at typical collision speeds and temperatures. Calculated melt thickness in collisions between ice spheres and a plane are given below. The thickness d_m is given in molecular layers , the sphere radius R in micrometers, and the approach speed V in m/s:

R = 10, V = 1, T = -35° C; d_m = 6;

R = 25, V = 3, T = -15° C, d_m = 40;

R = 50, V = 10 m/s, T = -5° C; d_m = 500.

From these values one sees that the melt thickness is very small at low velocities and temperatures. This provides a reason for the lower temperature limit of a thundercloud's active charging region

CONCLUSION

A theory is proposed to explain the collisional charging of ice, which is believed to underlie the process of electrification of thunderstorms. Based on clues obtained from many observations of storms and laboratory measurements, the theory describes the charging process on the molecular level. It explains the systematic dependence of charge and mass transfer between colliding particles, as functions of ambient temperature, relative growth rates and collision speeds. The theory requires only a single adjustable parameter to obtain quantitative agreement with laboratory measurements . However, the model is incomplete with respect to storms, in that several aspects of the collisional charging process are difficult to characterize. Further development of the theory, together with new measurements can improve the approximations, and new laboratory experiments can test its assumptions and lead to refinements.The theory should be extended to adapt it more directly to environmental parameters; so that it may eventually become a practical tool for the study and analysis of storm phenomena.

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REFERENCES

1. D.R. MacGorman and W.D. Rust, The Electrical Nature of Storms, (Oxford Univ. Press, N.Y)., 1998.

2. Saunders, C.P.R. Thunderstorm electrification laboratory experiments and charging mechanisms, J. Geophys.Res. 99, 10773-9 1994.

3. Mason, B.L. and J.G. Dash, Charge and mass transfer in ice-ice collisions: experimental observations of a mechanism in thunderstorm electrification, J.Geophys. Res.105, 10,185-10,192, 2000.

4. Dash, J.G., B.L.Mason and J.S.Wettlaufer, Theory of charge and mass transfer in ice-ice collisions, J.Geophys.Res., in press.

5. Smith, R.W. and D.J. Srolovitz, Void formation during film growth: A molecular dynamics simulation study, J. Appl. Phys. 79, 1448-57, 1996.

6. Einstein, A., On the movement of small particles suspended in a stationary liquid demanded by the molecular-kinetic theory of heat, in Investigations on the Theory of Brownian Movement. (Dover, New York) 1956, p. 1-18..

7. Verwey, E.J.W. and J.Th.G. Overbeek, Theory of Stability of Lyophobic Colloids, (Elsevier, Amsterdam, 1948).

8. Israelachvili, J.N., Intermolecular and Surface Forces, (Academic Press, London), Ch.12, 1992.

9. Dash, J.G. H.-Y. Fu, and J.S. Wettlaufer, The premelting of ice and its environmental consequences, Rep. Prog. Phys., 58, 115-67, 1995.

10. Baker, B., M.B. Baker, E.R. Jayaratne, J. Latham and C.P.R. Saunders, The influence of diffusional growth rates on the charge transfer acompanying rebounding collisions between ice crystals and soft hailstones, Q. J. R.Meteorol. Soc., 113, 1193-1215, 1987.

11. Caranti, G.M., E.E.Avila and M.A.Re, Charge transfer during individual collisions in ice growing from vapor deposition, J. Geophys. Res. 96, 15,365-15,375, 1991; Saunders, C.P.R. and S.L.Peck, Laboratory studies of the influence of the rime accretion rate on charge transfer during crystal/graupel collisions. J. Geophys. Res. 103, 13,949-13,961 1998; Keith, W.D. and C.P.R. Saunders, Further laboratory studies of the charging of graupel during ice crystal interactions, Atmos. Res. 25, 445-64, 1990.

12. Jayaratne, E.R., Charge separation during the impact of sand on ice and its relevance to theories of thunderstorm electrification, Atmos. Res., 26}, 407-24, 1991; Jayaratne, E.R., Density and surface temperature of graupel and the charge separation during ice crystal interactions, J. Geophys. Res. 103, 13,957-13,961, 1998.

13. Caranti, J.M. and A.J. Illingworth, The contact potential of rimed ice, J. Phys. Chem. 87, 4125-30, 1983.

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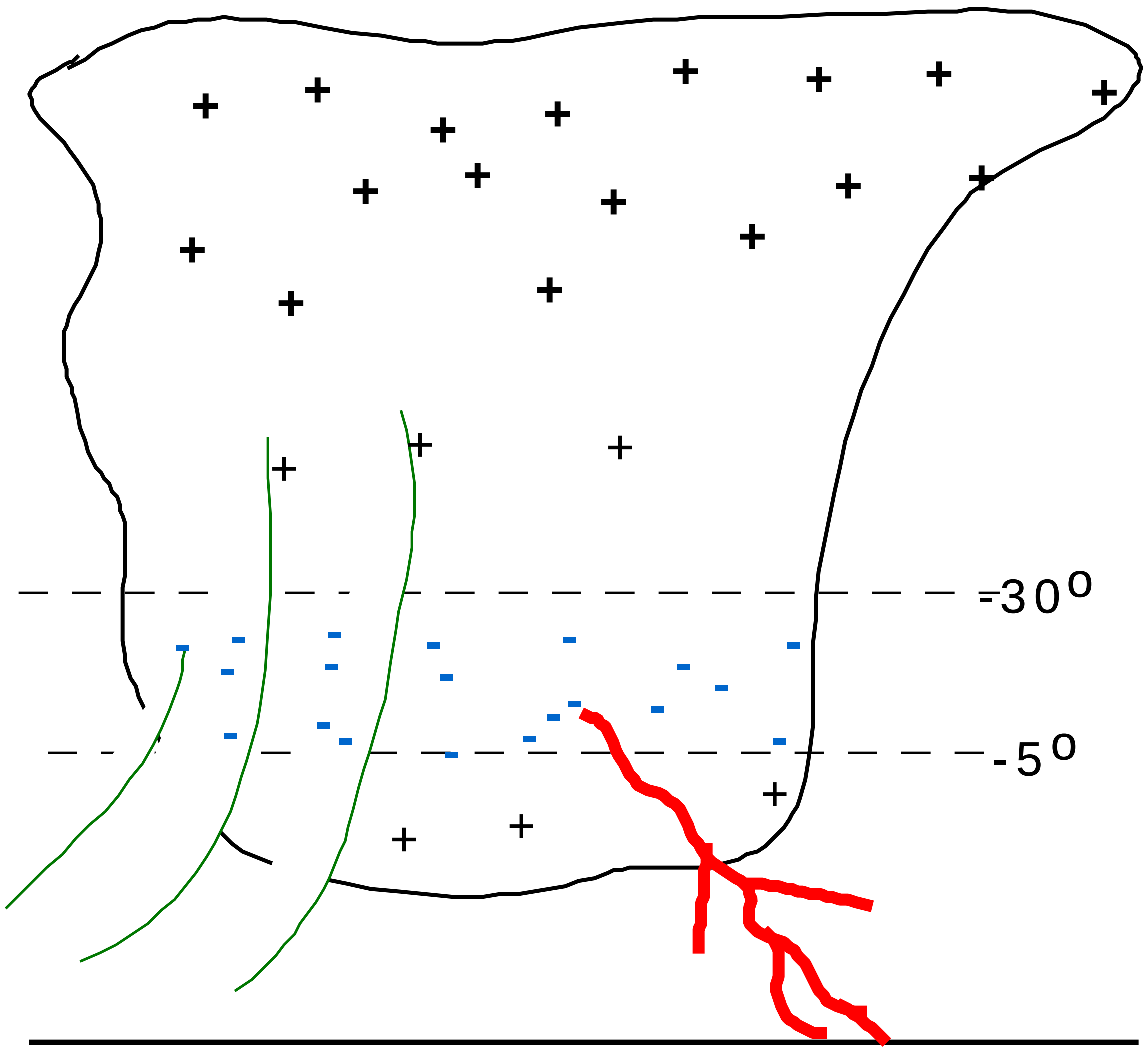

Fig.1a. Anatomy of a common thunderstorm, showing a principally dipolar charge distribution. Negative lightning-to-ground strokes originate in a region of active ice formation at temperatures between -5° and -30° containing negatively charged hail. Ice particles and unfrozen water droplets are carried upward by wind currents and are found throughout most of the cloud.

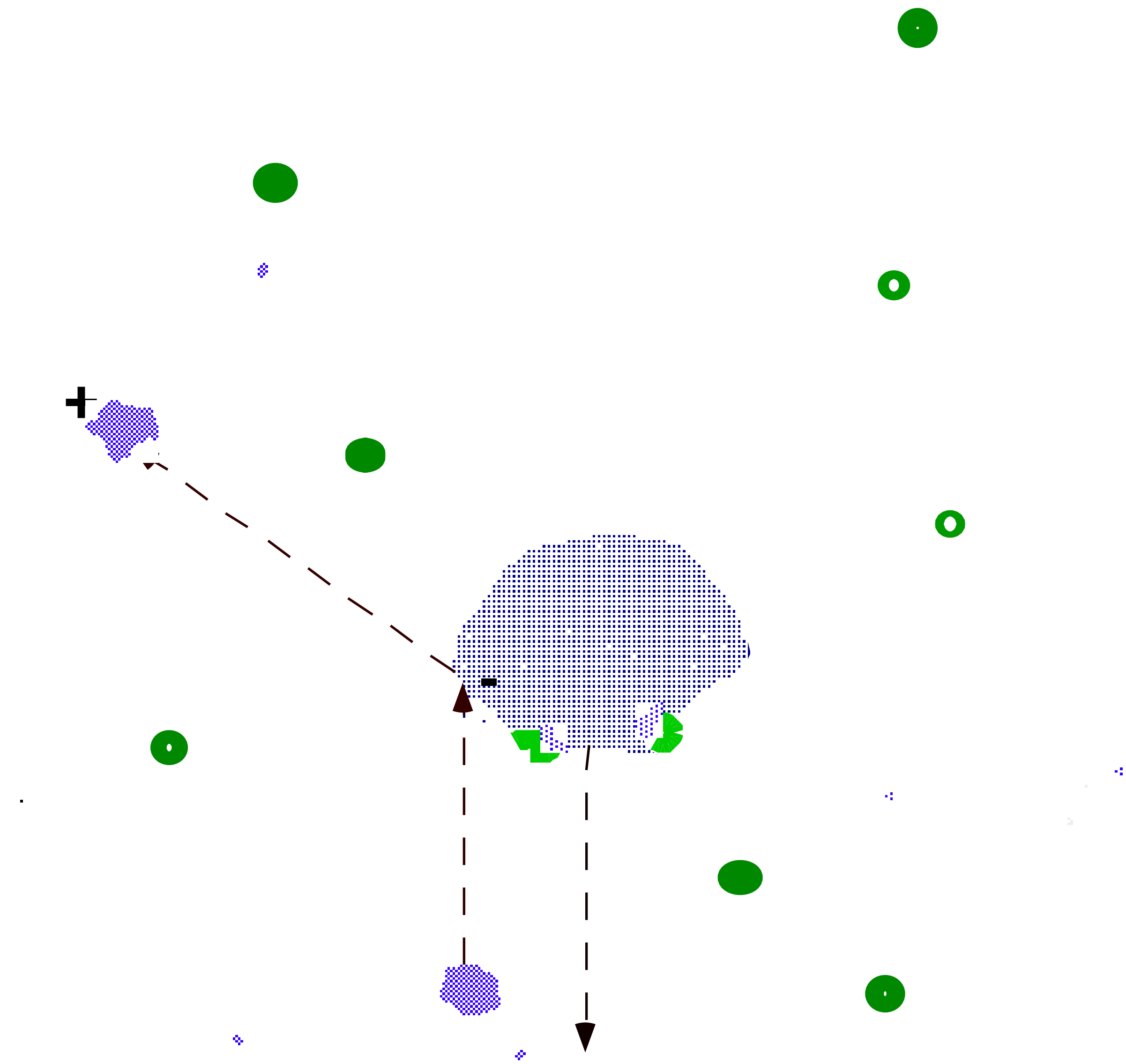

Fig.1b. Charging takes place in a region containing small ice particles rising in updrafts, falling graupel particles (soft hail), and supercooled liquid water droplets. Laboratory studiesindicate that reboundingcollisions between the ice particles and graupel, in the presence of superccoled water droplets, can account for the observed electric fields in lightning storms. The figure illustrates the dominant polarity resulting from ice-graupel collisions in storms with negative cloud-to-ground lightning.

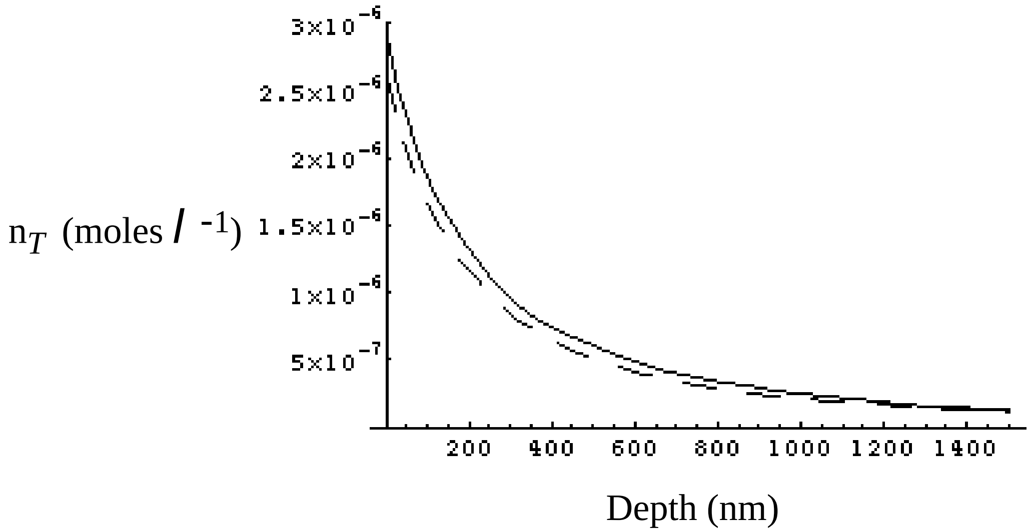

Fig.2a. Charge density versus depth from the surface. The ordinate is n_T, the net charge density, which is positive. The negative charge is localized at the surface. The curves are calculated for a surface charge density comparable to that of the MD experiments at temperatures between –5^{o}C and –20^{o}C. (4).

V(m s^{-1})

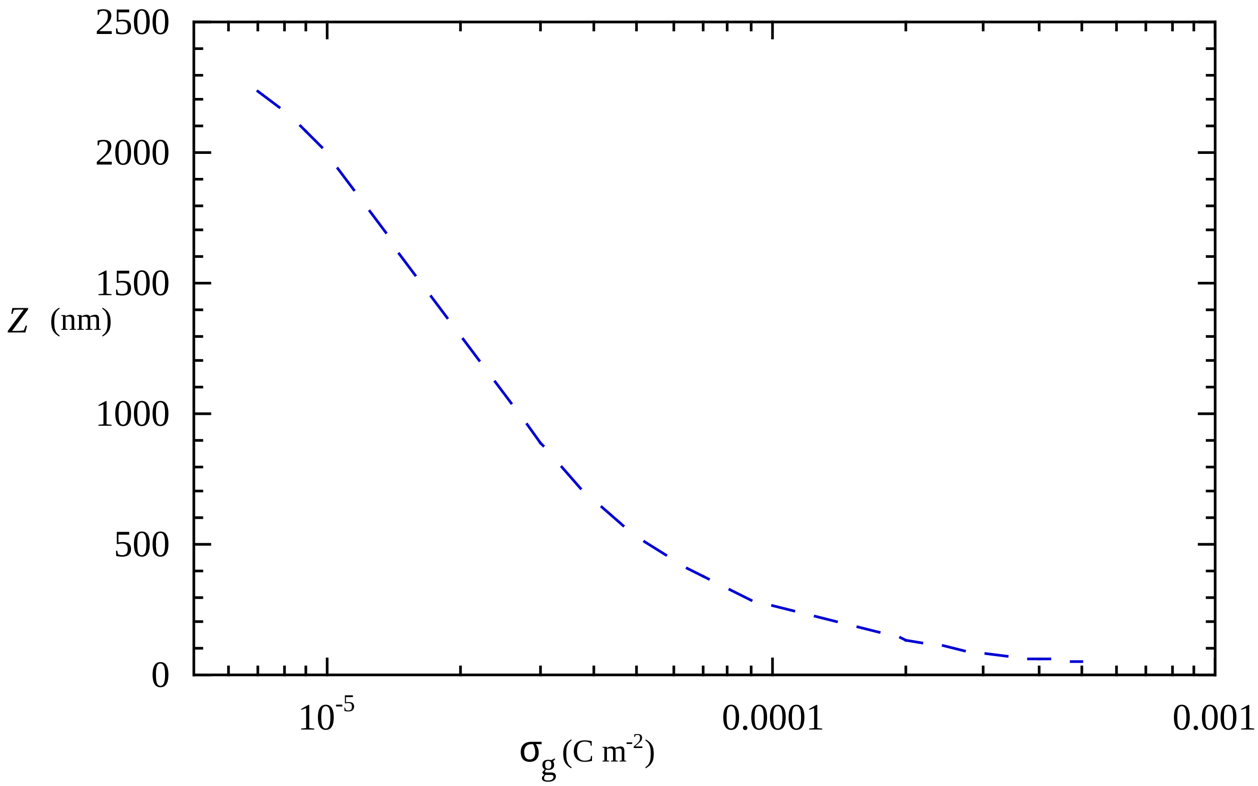

Fig. 2b The effect of growth rate Von the depth of the positive charge distribution. Z is the depth at which the net ion density n_T reaches 1/e of its value at the interface. The surface charge density σ_g is proportional to the growth rate (4).

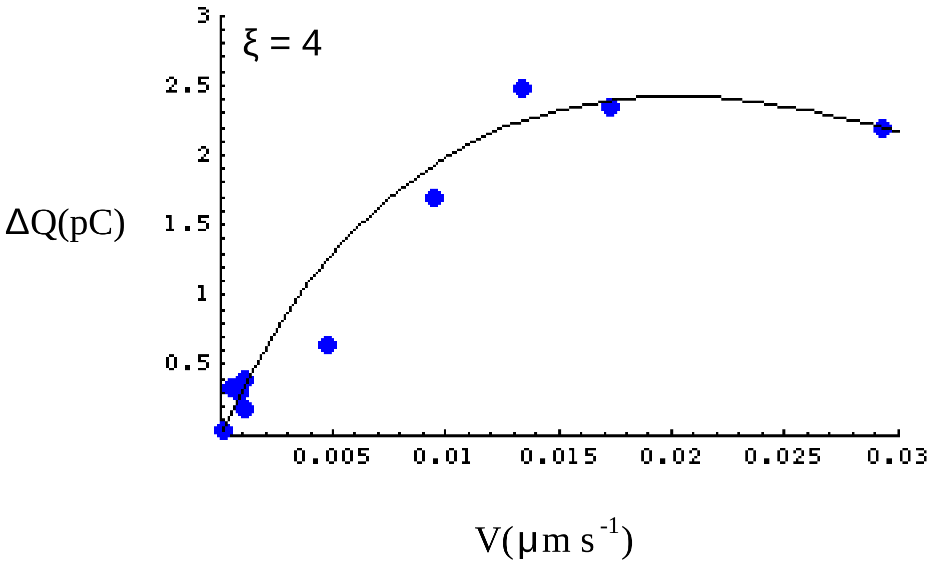

Fig. 3 Charge transfer versus growth rate calculated from the theory, compared with MD data. ΔQ is the charge over the collision area 10^{-8} m^2 of the experiment gained by the growing particle versus the growth rate over a range spanning the MD study. The theoretical curve is calculated with the experimental values of impact energy, collision area, and mean temperature, and a temperature difference of 3 K, greater than the 1 K estimated in the experiment. The surface structural disorder factor ξ = 4 yields the best fit overall. The saturation is driven by the neutralization provided by the dissolved cations. As more material participates in the collision, less negative charge is lost to the evaporating surface, and for a given total depth of shared melt liquid, the net negative charge loss decreases as the bulk ion density increases.