Coagulation of Charged Microparticles in Neutral Gas and Charge-Induced Gel Transitions

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Coagulation of charged particles was studied using the mean-field Smoluchowski equation. The coagulation equation was generalized for the case of a conserved system of charged particles. It was shown that runaway cluster growth (gelation) solutions exist if the charge-dipole (induced) interaction of clusters is included. When clusters are in thermal equilibrium with the ambient gas, the charge-dipole interaction dramatically enhances the aggregation process and considerably increases the likelihood of a gelation transition.

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Coagulation (or aggregation, clustering) is very important for various processes in different branches of physics and chemistry; for instance, polymerization [1], transitions in colloidal systems [2], plasma etching [3], planet formation [4], etc. In many cases coagulation can be considered as an irreversible process of cluster merging, due to the pair interaction of smaller clusters [5]. Different aspects of the aggregation theory for uncharged systems were studied in detail in the past two decades (see, e.g., [5–7] and references therein).

Experimental study of coagulation in a cloud of micron size particles embedded in a rarefied neutral gas is quite important for understanding the nature of the coagulation process [8]. During a recent series of the "PlasmaKristall-Experiment–Nefedov" experiments [9] performed onboard the International Space Station, we investigated the coagulation of micron size monodisperse particles in a neutral gas under microgravity [10]. In several experimental runs up to $\sim 10^6$ melamine-formaldehyde particles of 3.4 μm size were injected into the chamber filled with Ar gas at a pressure of 0.7 mbar. The mass spectrum of the resulting particle clusters (number of clusters n containing k particles) was measured. It was shown that starting from a certain moment $n(k)$ is not bounded at large k, but exhibits an algebraic tail, $n(k) \propto k^{-\tau}$. Simultaneously, the growth of a single large agglomerate occurs, accumulating $\sim 10^4$–10^5 particles in a fewseconds. The coagulation process develops several orders of magnitude faster than was expected. A huge agglomerate is formed while the aggregation among smaller clusters is still going on. All the observed features are peculiar to the so-called "gelation transition" (runaway cluster growth). This phenomenon is well known in the aggregation theory of uncharged systems [6,11].

Further investigation of the data obtained in the experiments [10] showed that the clusters were charged, positively or negatively. The charge magnitude was at least a few thousand electron charges. We believe that the enormously fast aggregation we observed is due to the charging. Kinetics of the charge-induced coagulation in neutral gases is very different from that in plasmas, where the charging is due to the absorption of electrons and ions and charge of the cluster is a certain function of the size (mass). In our case, however, the external sources of charging are absent and the total (initial) charge of the system is conserved.

In this Letter, we propose a theory of pair clustering in a *conservative* charged system. The simplest way to describe the aggregation process is to use the so-called mean-field theory and to generalize the Smoluchowski coagulation equation for the case of two independent variables—the cluster mass m and the charge Q. Then the kinetic (coagulation) equation for the distribution function of clusters $n(m, Q, t)$ can be written in the following form:

$$\frac{\partial}{\partial t} n(m, Q, t) = \frac{1}{2} \int_0^m dm' \int_{-\infty}^{\infty} dQ' K(m', Q'; m - m', Q - Q') n(m - m', Q - Q', t) n(m', Q', t) - n(m, Q, t) \int_0^{\infty} dm' \int_{-\infty}^{\infty} dQ' K(m', Q'; m, Q) n(m', Q', t), \tag{1}$$

where $K(m', Q'; m, Q)$ is the coagulation rate coefficient (kernel)—the probability for a pair of clusters, (m', Q') and (m, Q), to merge. The kernel is obviously symmetric with respect to the pair exchange: $K(m', Q'; m, Q) = K(m, Q; m', Q')$. The obtained equation is averaged over the velocity distribution, and the kernel is

$$K(m', Q'; m, Q) = \langle v_r \sigma(a', Q'; a, Q; v_r) \rangle, \tag{2}$$

where $v_r = |\mathbf{v} - \mathbf{v}'|$ is the relative velocity of the clusters and angle brackets denote averages. When clusters are in thermal equilibrium with an ambient gas, we have the

equipartition velocity dispersion, $\langle v_r \rangle \propto m_*^{-1/2}$, with $m_* = mm'/(m + m')$ the reduced mass of the pair. Below we consider this case. The merger cross section σ is a function of the "effective" aggregate radius a. The explicit relation between a and m is given by the appropriate scaling law with fractal dimension D_f [5],

$$m \propto a^{D_f}. \tag{3}$$

For different aggregation processes, the fractal dimension can vary from $D_f \simeq 3$ (dense, or compact clusters, upper limit) down to $\simeq 1.4$–1.5 (fluffy aggregates) [5,7].

Obtaining exact solutions of the coagulation equation is not a simple task even in the charge-independent case. The solutions with arbitrary initial conditions are known for only a few types of the kernel (e.g., $K = \text{const}$, $\propto m + m'$, and $\propto mm'$) [6]. The asymptotic solutions for the high-mass tail are also known for $K \propto m^\mu m'^\nu$ (with $\max\{\mu, \nu\} \leq 1$) [12]. For a constant kernel, the asymptotic solution was obtained for the distribution that depends on a vector of conserved quantities [13], which in the case of two components can be interpreted as the dependence on mass and charge. No analytic solutions are known for the charge-dependent kernels. However, the major features of the aggregation process can be understood by analyzing moments of the distribution function,

$$M_{\alpha,\beta}(t) = \int_0^\infty dm \int_{-\infty}^\infty dQ m^\alpha Q^\beta n(m, Q, t). \tag{4}$$

The general moments equations are derived by multiplying Eq. (1) with $m^\alpha Q^\beta$, integrating over m and Q, and changing the order of integration in the first integral. The result is

$$\dot{M}_{\alpha,\beta} = \frac{1}{2} \int_0^\infty \int_0^\infty dm' dm \int_{-\infty}^\infty \int_{-\infty}^\infty dQ' dQ [(m + m')^\alpha (Q + Q')^\beta - m^\alpha Q^\beta - m'^\alpha Q'^\beta] K(m', Q'; m, Q) n' n, \tag{5}$$

where $n' \equiv n(m', Q', t)$. Equation (5) is valid as long as n is exponentially bounded at the high-mass and high-charge end—then the integrals converge for arbitrary α and β (the coagulation kernel usually has algebraic asymptotics at large m and Q).

To determine the coagulation rate coefficient (2), one has to derive the merger cross section. We assume that the relative motion of clusters during the collision is ballistic (whereas the overall motion is diffusive). Then the cross section is defined by the effective potential energy of the pair cluster interaction, $U_{\text{eff}}(r, \rho) = \mathcal{E}_r(\rho/r)^2 + U_{\text{ch}}(r) + U_{\text{d}}(r)$, with ρ the relative impact parameter and $\mathcal{E}_r = \frac{1}{2} m_* v_r^2$ the kinetic energy of the relative motion. A major contribution to the coupling energy is provided by the charge-charge and charge-dipole (induced) interactions (U_{ch} and U_{d}, respectively) with the following radial dependencies [14]:

$$U_{\text{ch}}(r) = \frac{QQ'}{r}, \qquad U_{\text{d}}(r) \simeq -\alpha_{\text{d}} \left[\frac{a^3 Q'^2}{r^2(r^2 - a^2)} + \frac{a'^3 Q^2}{r^2(r^2 - a'^2)} \right], \tag{6}$$

where α_{d} is the coefficient of the dipole interaction. For a sphere of dielectric permittivity ϵ the coefficient is $\alpha_{\text{d}} = (\epsilon - 1)/(\epsilon + 2)$. The charge-charge interaction is stronger when *both* clusters are highly charged, so that the merger energy of the interaction, $|U_{\text{ch}}(a + a')|$, exceeds considerably the mean kinetic energy $\langle \mathcal{E}_r \rangle$. The charge-dipole interaction decreases with r much faster than the charge-charge interaction and, therefore, can only be important at $r \sim a + a'$. However, when one or both clusters are weakly charged and $|U_{\text{ch}}(a + a')| \lesssim \langle \mathcal{E}_r \rangle$, the range of the charge-charge interaction becomes shorter and the charge-dipole interaction is more important: One can show using Eq. (6) that $|U_{\text{d}}(a + a')|$ is normally larger than $|U_{\text{ch}}(a + a')|$ (unless both Q'/Q and a'/a lay in a certain relatively narrow range, or α_{d} is too small). Hence, in the first case ("highly charged clusters") the cross section is [15]

$$\sigma_{\text{ch}} = \pi(a + a')^2 (1 - \mathcal{E}_{\text{ch}}/\mathcal{E}_r), \tag{7}$$

with $\mathcal{E}_{\text{ch}} \equiv U_{\text{ch}}(a + a') = QQ'/(a + a')$ the merger energy. Note that, for clusters having the same sign of charge and $\mathcal{E}_{\text{ch}} > \mathcal{E}_r$, the cross section is equal to zero due to Coulomb repulsion. In the second case, when the clusters interact mostly via $U_{\text{d}}(r)$, the expression for the cross section cannot be derived in the general case. For $a \sim a'$ the coupling can be approximated by $U_{\text{d}}(r) \simeq -\alpha_{\text{d}}(a^3 Q'^2 + a'^3 Q^2)/r^4$, and the corresponding cross section is [15]

$$\sigma_{\text{d}} = \pi(a + a')^2 \times \begin{cases} 2\sqrt{\mathcal{E}_{\text{d}}/\mathcal{E}_r}, & \mathcal{E}_{\text{d}}/\mathcal{E}_r > 1, \\ (1 + \mathcal{E}_{\text{d}}/\mathcal{E}_r), & \mathcal{E}_{\text{d}}/\mathcal{E}_r < 1, \end{cases} \tag{8}$$

with $\mathcal{E}_{\text{d}} \equiv -U_{\text{d}}(a + a') = \alpha_{\text{d}}(a^3 Q'^2 + a'^3 Q^2)/(a + a')^4$ is the merger energy. In the absence of charges the aggregation is determined by the geometrical cross section, $\sigma_g = \pi(a + a')^2$.

Before solving the moment equations (5) with the kernels corresponding to the cross sections (7) and (8) let us outline some universal properties of Eq. (5) relevant to arbitrary kernels. As long as Eq. (5) is valid, the coagulation equation (1) provides the conservation of mass and charge. In terms of moments we get $\dot{M}_{1,0} = \dot{M}_{0,1} = 0$. Introducing the cluster number density as $M_{0,0} = N(t)$, we obtain from Eq. (5) that $\dot{N}(t)$ is always negative and thus the density decreases monotonically with time, as it should be for irreversible aggregation. The mass and charge conservation can be rewritten as $N\langle m \rangle = C_{1,0}$ and $N\langle Q \rangle = C_{0,1}$, with $\langle m \rangle$ and $\langle Q \rangle$ the

average mass and charge per cluster, which increase with time.

We make the following assumption about the initial conditions, which simplifies significantly the subsequent analysis: Suppose that the initial overall charge of the particles equals zero, i.e., $C_{0,1} = 0$. If we assume further that the initial charge distribution is symmetric, $n(m, -Q, 0) = n(m, Q, 0)$, then the symmetry is conserved, since the coagulation rate is an even function of the charges: $K(m', -Q'; m, -Q) = K(m', Q'; m, Q)$. Then we get from Eqs. (4) and (5) that all odd Q moments are identically equal to zero, and even moments ($\beta = 2i$) are $M_{\alpha,\beta} = 2\int_0^\infty dm \int_0^\infty dQ m^\alpha Q^\beta n$. Therefore, the odd Q moments ($\beta = 2i + 1$) we use below are the "absolute charge" moments: $M_{\alpha,\beta}(m, |Q|) \equiv 2\int_0^\infty dm \int_0^\infty dQ m^\alpha |Q|^\beta n$.

Let us now study how much the charge-induced interaction can enhance the aggregation.

Charge-charge interaction.—Assuming that the merger energy of the interaction exceeds significantly the average kinetic energy, $\mathcal{E}_{\rm ch} \gg \langle \mathcal{E}_{\rm r} \rangle$, we get from Eq. (7) the following expression for the coagulation rate:

$$K(m', Q'; m, Q) \simeq \begin{cases} -K_0(m', m)Q'Q, & Q'Q < 0, \\ 0, & Q'Q > 0, \end{cases} \quad (9)$$

where $K_0 = \pi(a + a')\langle v_{\rm r}/\mathcal{E}_{\rm r} \rangle$. Coagulation between oppositely charged clusters is possible only in this limit, and thus the charge distribution becomes narrower with time. The coagulation is enhanced compared to the uncharged case by the factor $\sim \mathcal{E}_{\rm ch}/\langle \mathcal{E}_{\rm r} \rangle$. In order to demonstrate general features of the solution of the moment equations, let us assume $K_0(m', m) = \text{const}$. This does not change the results qualitatively, but makes them much simpler (unless K_0 increases too steeply with m and m', which is not the case). Substituting Eq. (9) into the moment equations (5), we obtain for the second charge and mass moments:

$$\begin{aligned} \dot{M}_{0,2} &= -\tfrac{1}{2} K_0 M_{0,2}^2, \\ \dot{M}_{2,0} &= \tfrac{1}{2} K_0 M_{1,1}^2. \end{aligned} \quad (10)$$

From the first Eq. (10) we get the charge dispersion: $M_{0,2}(t) \equiv N\langle Q^2 \rangle = M_{0,2}^0 (1 + \frac{1}{2} K_0 M_{0,2}^0 t)^{-1}$, i.e., it decreases as $\propto t^{-1}$ at large time scales. The mean cluster mass $s(t)$ is determined by the mass dispersion: $s = M_{2,0}/M_{1,0}$. Using the second Eq. (10) we can easily evaluate the upper bound for $M_{2,0}(t)$. From the Cauchy inequality we obtain $\dot{M}_{2,0} \le \frac{1}{2} K_0 M_{2,0} M_{0,2}$. Substituting $M_{0,2} \simeq (\frac{1}{2} K_0 t)^{-1}$, we have $\dot{M}_{2,0} \le M_{2,0}/t$. Hence, $M_{2,0} \propto t$, i.e., the mean cluster mass does not grow faster than linearly.

Charge-dipole interaction.—The charge dispersion decreases with time due to the charge-charge enhanced coagulation, and when the mean merger energy of the charge-charge interaction ($\propto \langle Q^2 \rangle$) becomes less than $\langle \mathcal{E}_{\rm r} \rangle$, the charge-dipole interaction is more important. The corresponding coagulation rate is a function of the absolute values of the charges, $K = K(m', |Q'|; m, |Q|)$. For this type of the kernel there exists one more integral of the coagulation equation: $\dot{M}_{0,2} = 0$, or $N\langle Q^2 \rangle = C_{0,2}$. Since N always decreases with time, the charge dispersion grows, in contrast to the case of the charge-charge interaction. We write the coagulation kernel in the following algebraic form (omitting a constant factor):

$$K(m', Q'; m, Q) = m^\mu m'^\nu |Q'|^\varepsilon + m'^\mu m^\nu |Q|^\varepsilon, \quad (11)$$

where μ and ν are the mass exponents ($\mu + \nu \equiv \lambda$), and ε the charge exponent. (For the sake of convenience, we also omit the part corresponding to the geometric cross section; its role is discussed in the conclusion.) The values of μ and ν can be obtained from the asymptotic behavior of the general expression for K in the limit $a/a' \gg 1$ [6]. However, the expression for the charge-dipole coagulation cross section and thus for the corresponding kernel can be derived analytically only for $a/a' \sim 1$ [see Eq. (8)]. Nevertheless, we apply the cross section (8) for the analysis below: This expression was obtained using the approximation for the interaction energy, which (the absolute value) is always smaller than the actual $|U_{\rm d}(r)|$ from Eq. (6). Hence, Eq. (8) allows us to estimate the *lower edge* of the charge-dipole coagulation rate. This gives the charge exponent equal to 1 or 2, depending on the ratio $\mathcal{E}_{\rm d}/\langle \mathcal{E}_{\rm r} \rangle$ and, using the scaling law (3), yields the total mass exponent $\lambda = \frac{3}{2} D_{\rm f}^{-1} - \frac{1}{2}$ for $\varepsilon = 1$ and $\lambda = D_{\rm f}^{-1} - \frac{1}{2}$ for $\varepsilon = 2$.

It is well known that in some uncharged systems a special kind of phase transition called "gelation" is possible (see, e.g., [6,11] and references therein). At a certain "gelation moment" $t_{\rm gel}$ the ensemble becomes unstable against the formation of a single cluster of "infinite" mass. This process is also called "runaway growth." The gelation develops if the coagulation rate increases sufficiently steeply with the mass, $\lambda > 1$. Mathematically, this is because at $t = t_{\rm gel}$ the distribution function for such kernels is no longer bounded exponentially at the high-mass end, but behaves algebraically, $n(m, t_{\rm gel}) \propto m^{-\tau}$ with $2 < \tau < 3$. In terms of the moments it means that the gel particle accumulates the mass comparable with the total mass of the system, so that the mass dispersion, or the mean cluster mass, $s(t_{\rm gel}) \propto M_2(t_{\rm gel})$, diverges. Let us show that the aggregation process with the charge-dipole interaction can enhance the gelation dramatically. As an example, consider the coagulation kernel of the form: $K(m', Q'; m, Q) = mQ'^2 + m'Q^2$. For this kernel $\lambda = 1$ and thus gelation without the charge-induced interaction (corresponding to $\varepsilon = 0$) is not possible. For the cluster density we have $\dot{M}_{0,0} = -C_{1,0} C_{0,2}$, i.e., the density decreases linearly, $N(t) = N^0(1 - t/t_N)$, where $t_N = N^0/(C_{1,0} C_{0,2})$. For the higher moments we have

$$\begin{aligned} \dot{M}_{2,0} &= 2M_{1,2}M_{2,0}, \\ \dot{M}_{1,2} &= M_{1,2}^2 + M_{2,0}M_{0,4}, \\ \dot{M}_{0,4} &= 6M_{1,2}M_{0,4}. \end{aligned} \tag{12}$$

Using the first and the third equations we get (neglecting lower order terms) $\dot{M}_{1,2} \simeq 4M_{1,2}^2$. The solution of this equation has a singularity, $M_{1,2}(t) \propto (1 - 4M_{1,2}^0 t)^{-1}$, the mean cluster mass diverges as well, $s(t) = M_{2,0}/M_{1,0} \propto M_{1,2}^{1/2}$, which is an indication of the gelation transition. Since $M_{1,2}^0 = C_{1,0}C_{0,2}/N^0 \equiv t_N^{-1}$, we get the gelation time $t_{\rm gel} = \frac{1}{4}t_N$ and the number density in this moment is $N(t_{\rm gel}) = \frac{3}{4}N^0$. Thus, we have shown that the charge-dependent coagulation can cause runaway growth. Note that $N\langle Q^2\rangle = {\rm const}$, and the charge dispersion does not change considerably up to the gelation moment $\langle Q^2\rangle|_{t=t_{\rm gel}} = \frac{4}{3}\langle Q^2\rangle|_{t=0}$. Therefore, the assumption that the charge-dipole interaction prevails is valid at least up to $t = t_{\rm gel}$.

Now we can derive the gelation transition criterion for the kernel (11). First we consider the case $\varepsilon = 1$. The behavior of $s(t)$ is determined by the equation $\dot{M}_{2,0} = 2M_{\mu+1,0}M_{\nu+1,1}$. Using the Hölder inequality, we can evaluate the *upper bound* for $M_{2,0}$ and thus find the *necessary* condition for the transition. First, we have $M_{\mu+1,0} \le C_{1,0}^{1-\mu}M_{2,0}^{\mu}$, which is valid for $0 < \mu < 1$. Then, $M_{\nu+1,1} \le C_{0,2}^{1/2}M_{2\nu+2,0}^{1/2}$, and transforming $M_{2\nu+2,0}$ (valid for $-1/2 < \nu < 0$) we obtain $\dot{M}_{2,0} \le 2C_{1,0}^{1-\lambda}C_{0,2}^{1/2}M_{2,0}^{\lambda+1/2}$. We see that the mean cluster mass diverges when $\lambda + \frac{1}{2} > 1$, i.e., the necessary condition for the gelation is $\lambda > \frac{1}{2}$.

Similarly, for $\varepsilon = 2$ we get $\dot{M}_{2,0} = 2M_{\mu+1,0}M_{\nu+1,2}$. Transforming $M_{\nu+1,2} \ge C_{1,0}^{-1}M_{\nu/2+1,1}^2$ and using the Hölder inequality for $M_{\nu/2+1,1}^2 \le C_{0,2}M_{\nu+2,0}$ (valid for $-1 < \nu < 0$), we derive $\dot{M}_{2,0} \propto C_{1,0}^{-\lambda}C_{0,2}M_{2,0}^{\lambda+1}$. Now, the gelation condition is $\lambda > 0$. Combining the obtained results we have the following gelation condition for the kernel (11):

$$\lambda + \tfrac{1}{2}\varepsilon > 1, \tag{13}$$

in contrast to $\lambda > 1$ for the charge-free case. Criterion (13) shows that the charge-dipole interaction enhances the aggregation significantly and stimulates the gel phase transition.

Let us compare the gelation conditions for the charge-dipole and pure geometrical coagulation. Equation (13) shows that in the presence of charge-induced interactions the kernel need not necessarily be a steep function of mass—for the charge exponent $\varepsilon = 2$ the mass exponent λ is sufficient to be positive. Smaller λ implies higher values of the fractal dimension $D_{\rm f}$ of clusters. Using Eq. (13) we get the critical $D_{\rm f}$ which are necessary to start the gelation for different types of coagulation: For charge-dipole coagulation with $\varepsilon = 2$ we have $\lambda = D_{\rm f}^{-1} - \frac{1}{2}$, and the gelation condition is $D_{\rm f} < 2$; with $\varepsilon = 1$ we have $\lambda = \frac{3}{2}D_{\rm f}^{-1} - \frac{1}{2}$, and $D_{\rm f} < \frac{3}{2}$. For geometrical coagulation $\lambda = 2D_{\rm f}^{-1} - \frac{1}{2}$, and the gelation at $D_{\rm f} < \frac{4}{3}$.

These results are obtained for the case of (i) equipartition (thermal) velocity distribution, when $\langle v_{\rm r}\rangle \propto m_*^{-1/2}$ ($\langle \mathcal{E}_{\rm r}\rangle = {\rm const}$). It is worthwhile to consider the opposite limit of (ii) mass-independent velocity dispersion, with $\langle v_{\rm r}\rangle = {\rm const}$ ($\langle \mathcal{E}_{\rm r}\rangle \propto m_*$). The values of λ are different in this case: For charge-dipole coagulation with $\varepsilon = 2$, we have $\lambda = D_{\rm f}^{-1} - 1$, and the gelation when $D_{\rm f} < 1$; with $\varepsilon = 1$ we have $\lambda = \frac{3}{2}D_{\rm f}^{-1} - \frac{1}{2}$, and $D_{\rm f} < \frac{3}{2}$. For geometrical coagulation $\lambda = 2D_{\rm f}^{-1}$, and the gelation at $D_{\rm f} < 2$.

Clusters produced due to the Brownian motion are very fragile and fluffy, with an average value of the fractal dimension about $\simeq 1.8$–2 [7]. Thus, for the velocity distribution (i), e.g., when clusters are in thermal equilibrium with ambient gas, the gelation is more probable due to the charge-dipole interaction. In contrast, for the distribution (ii), e.g., for a beam of "cold" particles, the clusters are rather dense ($D_{\rm f}$ is higher) and a gel transition is only possible for pure geometrical coagulation.

In conclusion, we generalized the Smoluchowski coagulation equation for conserved systems of charged particles. Analysis shows that the equation allows the runaway, or gelation, solution. When clusters are in thermal equilibrium, the charge-induced interaction dramatically enhances the aggregation process and makes the gel transition more likely.

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