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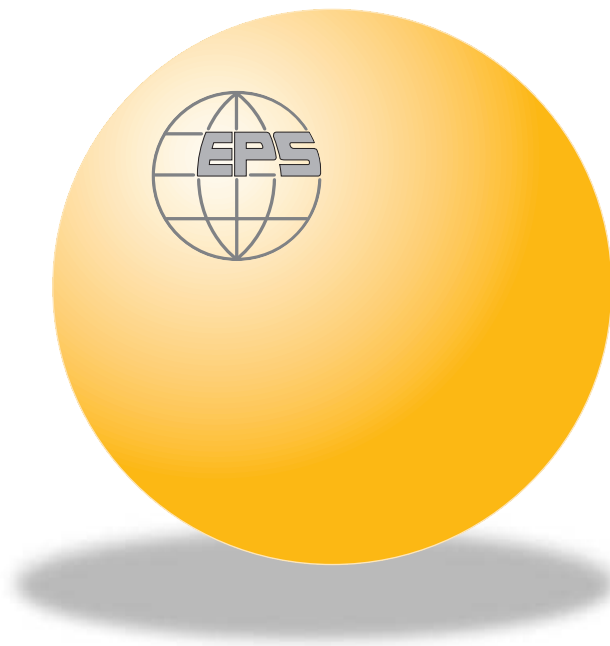
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Vol. 67 • Number 4 • pp. 545–551

## **Kinetic description of coagulation and fragmentation in dilute granular particle ensembles**

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F. SPAHN, N. ALBERS, M. SREMČEVIĆ and C. THORNTON



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# EUROPHYSICS LETTERS

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24 issues - Vol. 65-68 (6 issues per vol.)

ISSN: 0295-5075 - ISSN electronic: 1286-4854

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## Kinetic description of coagulation and fragmentation in dilute granular particle ensembles

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(received 16 December 2003; accepted in final form 4 June 2004)

PACS. 05.20.Dd – Kinetic theory.

PACS. 45.70.-n – Granular systems.

PACS. 96.35.Cp – Origin, formation, evolution, and ages.

**Abstract.** – We derive kinetic equations covering coagulation and fragmentation of granular gases including a combined dynamics of the mass spectrum and the velocity distribution. We will focus on coagulation, that can only occur at low impact velocities where attractive forces and dissipation prevent a post-collisional separation. We calculate an impact speed-dependent threshold velocity  $g_c$  for coagulation to occur based on binary collision dynamics of viscoelastic granular particles including adhesive forces and determined by the masses, and the material of the colliding particles. Growth processes are immensely slowed down due to  $g_c$  and the resulting restriction in phase space, and do furthermore depend on the ratio of threshold and thermal velocity of a considered particle ensemble. The Smoluchowski equation emerges from the general kinetic approach as a special case.

*Introduction.* – The evolution of dilute granular gases in time is determined by their spatial, velocity and mass distributions, and is dominated by particle collisions which can either result in i) coagulation, ii) restitution or iii) fragmentation. The granular gases are usually governed by dissipative non-destructive collisions, the regime termed here as restitution. Coagulation becomes possible if attractive forces are present allowing particles to coalesce, whereas fragmentation occurs if relative or thermal velocities of the ensemble are large enough to erode the particles. The predominance of either the coagulation, the restitution, or the fragmentation regime can vary strongly for different systems and determines the width of the respective domain, roughly sketched in fig. 1. In case of dry granular matter, where the number of colliding particles is conserved, kinetic theory has widely been used to treat the spatial and velocity distribution dynamics in terms of Boltzmann or Chapman-Enskog equations [1–4]. If the total mass of a system is conserved, but the mass distribution itself is able to change, fragmentation equations on the one hand and coagulation equations (generally based on the Smoluchowski equation [5]) on the other hand have been applied. The interplay between coagulation and fragmentation is important in many fields, but corresponding integro-differential equations mainly focus on the mass distribution only [6–9]. Some authors have also incorporated a restricted coagulation domain (fig. 1) resulting in a modified Smoluchowski equation [10, 11]. Many authors introduced “hybrid” models, where the mass evolution is treated with a kinetic-like equation (fragmentation or coagulation), while

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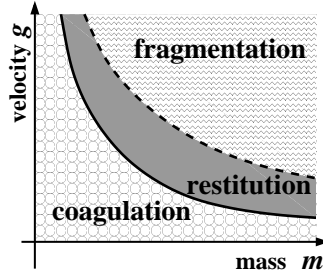


Fig. 1 – Domain diagram: mass  $m$  and relative impact speed  $g$  determine the physical scenario.

the velocity and positional evolution is considered separately (particle in a box or similar approximations; *e.g.*, [12–14]).

In this letter we introduce a self-consistent treatment of *mass*, *spatial* and *velocity distribution*. The derived kinetic equation is then applied to the coagulation case under simplifying assumptions for the velocity distribution, giving a modified coagulation equation. The coagulation domain (fig. 1) will be obtained by analyzing binary collision dynamics including adhesion and dissipation. The Smoluchowski equation will be derived and appears to be a special case valid under certain constraints. Finally, we will demonstrate differences arising from the restricted coagulation domain in a numerical example.

*Kinetic equations.* – Denoting the particle's mass  $M$ , location vector  $\mathbf{R}$ , velocity  $\mathbf{V}$  and the time  $t$ , we introduce a generalized distribution function  $F(M, \mathbf{R}, \mathbf{V}, t)$ .  $F$  determines the number of particles  $dN$  in the volume  $d^3\mathbf{V} d^3\mathbf{R} dM$  by  $dN = F(M, \mathbf{R}, \mathbf{V}, t) d^3\mathbf{R} d^3\mathbf{V} dM$ . Referring to a local area in phase space, the time evolution of  $F$  is given by a rate equation between gain  $\mathcal{G}^{(c/f)}$  terms and loss  $\mathcal{L}^{(c/f)}$  terms corresponding to coagulation (c) and fragmentation (f):

$$\frac{D}{Dt} F(M, \mathbf{R}, \mathbf{V}, t) = \left. \frac{DF}{Dt} \right|^{(c)} + \left. \frac{DF}{Dt} \right|^{(f)} = \mathcal{G}^{(c)} - \mathcal{L}^{(c)} + \mathcal{G}^{(f)} - \mathcal{L}^{(f)}. \quad (1)$$

The substantial derivative is denoted by  $D/Dt$  accounting for the system dynamics except for binary particle interactions. Assuming all particles to be independent apart from collisions, thus separating  $F^{(2)}(m, \mathbf{r}, \mathbf{v}, t; m_1, \mathbf{r}_1, \mathbf{v}_1, t) = F(m, \mathbf{r}, \mathbf{v}, t) \cdot F_1(m_1, \mathbf{r}_1, \mathbf{v}_1, t)$ , and changing to a centre-of-mass frame with the relative  $\mathbf{g} = \mathbf{v}_1 - \mathbf{v}$  and the centre-of-mass velocity  $\mathbf{v}_s = (m\mathbf{v} + m_1\mathbf{v}_1)/(m + m_1)$ , the gain and loss terms can be written as

$$\mathcal{G}^{(c/f)}(M, \mathbf{R}, \mathbf{V}, t) = \int_0^\infty dm \int_0^\infty dm_1 \int_{\mathbf{R}^3} d^3\mathbf{v}_s \int_{\mathcal{D}_{c/f}} d^3\mathbf{g} W_{\mathcal{G}}^{(c/f)}(M, \mathbf{V}; m, m_1, \mathbf{g}) F F_1, \quad (2)$$

$$\mathcal{L}^{(c/f)}(M, \mathbf{R}, \mathbf{V}, t) = \int_0^\infty dm \int_0^\infty dm_1 \int_{\mathbf{R}^3} d^3\mathbf{v}_s \int_{\mathcal{D}_{c/f}} d^3\mathbf{g} W_{\mathcal{L}}^{(c/f)}(M, \mathbf{V}; m, m_1, \mathbf{g}) F F_1. \quad (3)$$

The integrands on the RHS refer to the state prior to collision. Coagulation and fragmentation occur for impact speeds of  $0 < g < g_c(m, m_1)$  and  $g_f(m, m_1) < g < \infty$ , respectively.  $\mathcal{D}_{(c/f)}$  and  $W_{\mathcal{G}/\mathcal{L}}^{(c/f)}$  denote the corresponding integration domain and integral kernel. Since only approaching particles collide,  $\mathcal{D}_{(c)}$  reduces to a hemisphere in spherical coordinates and  $\mathcal{D}_{(f)}$  to velocity space minus an excluded hemisphere. The geometrical cross-section  $\sigma(m, m_1, \varrho) = \pi(3/(4\pi\varrho_p))^{2/3}(m^{1/3} + m_1^{1/3})^2$  is common to all domains and can be increased or even decreased by long-range particle interactions as there are, for instance, Coulomb

forces [15] or gravitational interactions [16]. These can be incorporated by a factor  $\mathcal{F}$ , including, *e.g.*, the charge of grains, their mass, or other particle properties. The mean particle mass density is denoted by  $\varrho_p$ . Hence, the integration kernels read in detail

$$W_{\mathcal{G}/\mathcal{L}}^{(c)} = \sigma |\mathbf{g}| \mathcal{F} \delta(m_{\mathcal{G}/\mathcal{L}} - M) \delta(\mathbf{v}_{\mathcal{G}/\mathcal{L}} - \mathbf{V}), \quad (4)$$

$$W_{\mathcal{G}}^{(f)} - W_{\mathcal{L}}^{(f)} = W^{(f)} = \sigma |\mathbf{g}| \mathcal{F} [\mathcal{P}(M, \mathbf{V} | m, m_1, \mathbf{g}) - \delta(m_1 - M) \delta(\mathbf{v}_1 - \mathbf{V})], \quad (5)$$

where  $m_{\mathcal{G}} = m + m_1$  and  $m_{\mathcal{L}} = m_1$ , and  $\mathbf{v}_{\mathcal{G}} = (m\mathbf{v} + m_1\mathbf{v}_1)/(m + m_1)$  and  $\mathbf{v}_{\mathcal{L}} = \mathbf{v}_1$ . These distinctions arise from different integration paths for gain ( $m + m_1 = \text{const}$ ) and loss ( $m = \text{const}$ ) terms in the  $(m, m_1)$  parameter space. Dirac's delta-function is denoted by  $\delta(x)$  and assures conservation of mass and momentum. Coagulating particles will form a *single* aggregate with a mass  $m + m_1$  and velocity  $\mathbf{v}_s$ . By assuming solid spherical particles  $m \propto R^3$  that do, even if they fragment or coagulate, form new solid spherical particles of different mass, we deliberately neglect fractal growth (agglomeration) and the impact geometry for simplicity. Nevertheless, this does neither qualitatively affect results given below nor change the dependences in eqs. (4)-(5), and can be implemented in more detailed studies accounting for generally observed growth of fractal shaped bodies [17] in form of  $m \propto R^{D_f}$ .

In case of destructive impacts, particles are destroyed forming post-collisional size and velocity distributions (eq. (5)). In contrast to coagulation, resulting fragments may cover a broad range of masses and velocities which are hard to derive from basic principles but can be gained from extensive experimental studies of fragmentation [18]. Bearing this in mind, the gain and loss term of fragmentation may be written using a normalized conditional distribution function  $\mathcal{P}(M, \mathbf{V} | m, m_1, \mathbf{g})$  of the generated debris as in eq. (5) and as suggested in [14]. Hence, the basic kinetic balance (1) becomes

$$\frac{D}{Dt} F(M, \mathbf{R}, \mathbf{V}, t) = \int_0^\infty dm \int_0^\infty dm_1 \int_{\mathbf{R}^3} d^3 \mathbf{v}_s \left[ \int_{\mathcal{D}_c} d^3 \mathbf{g} \left( \frac{1}{2} W_{\mathcal{G}}^{(c)} - W_{\mathcal{L}}^{(c)} \right) F F_1 + \int_{\mathcal{D}_f} d^3 \mathbf{g} W^{(f)} F F_1 \right]. \quad (6)$$

Due to symmetry with respect to an exchange of particles a factor of 1/2 appears in the coagulation gain term.

*Coagulation equation.* – Here, we will show the effects of the reduced coagulation domain  $\mathcal{D}_c$  in terms of a simple example. In favour of a coagulation equation, we neglect fragmentation and consider a force-free system ( $D/Dt \rightarrow \partial/\partial t$ ). To gain the size distribution dynamics, it is necessary to integrate eq. (6) with respect to  $\mathbf{V}$  (zeroth-moment balance). Without further detail about the distribution functions themselves, it will be complicated to proceed analytically. Thus, we assume a Maxwellian velocity dispersion characterized by energy equipartition  $v = (2T/m)^{1/2}$  [15] ( $T$  is the granular temperature). Additionally, we factorize the generalized distribution function  $F(m, \mathbf{r}, \mathbf{v}, t) = n(m, \mathbf{r}, t) \cdot f(m, \mathbf{v})$ , implying an independence of mass and velocity spectra. Although we are aware of the restrictions brought about by this approach, they will prove to be necessary assumptions for deriving the Smoluchowski equation. Bearing in mind that only a fraction of the velocity-mass space  $\mathcal{D}_c$  accounts for coagulation, we obtain the following coagulation equation:

$$\begin{aligned} \partial_t n(M, \mathbf{R}, t) = & \frac{1}{2} \int_0^M dm \mathcal{K}(m, M - m) n(m, \mathbf{r}, t) n(M - m, \mathbf{r}, t) - \\ & - n(M, \mathbf{r}, t) \int_0^\infty dm \mathcal{K}(m, M) n(m, \mathbf{r}, t), \end{aligned} \quad (7)$$

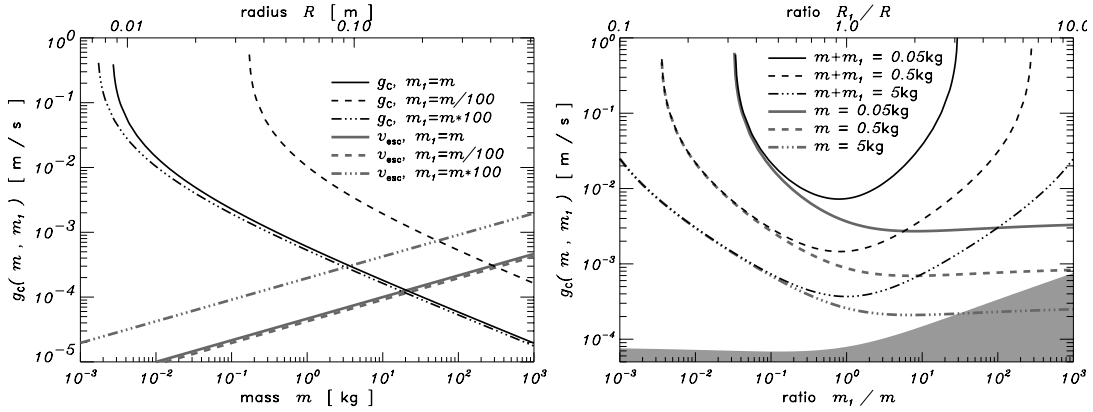


Fig. 2 – The dependence of the threshold impact speed  $g_c(m, m_1)$  evaluated for icy grains. Left: the dependence on  $m$  is presented for different  $\mu = m_1/m$ . The decreasing curves (black) are the numerical solutions of eq. (10). The grey straight lines show the corresponding gravitational escape velocities (12). Right: the dependence on  $\mu$  for different initial and final masses  $m$  and  $m + m_1$  is shown. The grey zone corresponds to the range where gravity (escape velocity  $v_{\text{esc}}$ ) dominates.

where

$$\mathcal{K}(m, m_1) = \frac{\sigma(m, m_1)}{\sqrt{\pi}} v_{\text{th}}(m, m_1) \mathcal{C}(g_c, v_{\text{th}}(m, m_1)). \quad (8)$$

The parameter  $v_{\text{th}}^2(m, m_1) = 2(m + m_1)T/(mm_1)$  represents an effective thermal velocity. The integration kernel is completed by

$$\mathcal{C}(g_c, v_{\text{th}}) = 1 - \left(1 + \frac{g_c^2}{v_{\text{th}}^2}\right) \exp\left[-\frac{g_c^2}{v_{\text{th}}^2}\right]. \quad (9)$$

In the limits of  $g_c \rightarrow \infty$  coagulation always occurs ( $\mathcal{C} \rightarrow 1$ ), or for  $g_c \rightarrow 0$  it cannot take place ( $\mathcal{C} \rightarrow 0$ ) at all. Equation (9) refers to a reduced coagulation which is dependent on the granular temperature and material properties of the considered ensemble. Within a narrow range of  $g_c/v_{\text{th}}$ ,  $\mathcal{C}(g_c, v_{\text{th}})$  changes its value from zero to one denoting a rather sharp boundary. It is furthermore remarkable that in case of  $\mathcal{C} \rightarrow 1$  ( $g_c \rightarrow \infty$ ) the otherwise phenomenological Smoluchowski equation is reproduced. Hence, the derivation of the Smoluchowski equation implies assumptions that should always be considered while application, as there are an *independence between mass and velocity spectrum*, *energy equipartition* and the *sticking* of particles whenever they collide.

*Coagulation domain and collision dynamics.* – The coagulation domain border  $g_c$  can be obtained by collision dynamics, where the binary contact itself is based on a viscoelastic collision model [19] taking into account adhesive forces [20]. For simplicity, we neglected rotations from the very beginning and thus the deformation at contact is defined as  $\xi(t) = R + R_1 - |\mathbf{r}(t) - \mathbf{r}_1(t)|$  yielding a differential equation for the normal component of the collision dynamics:

$$\ddot{\xi} + \frac{H}{m_{\text{eff}}} \left[ \xi^{3/2} + \frac{3}{2} A \dot{\xi} \xi^{1/2} - 3 \left( \frac{\pi \gamma_{\text{eff}}}{2 E_{\text{eff}}} \right)^{1/2} (R_{\text{eff}} \xi^3)^{1/4} \right] = 0, \quad (10)$$

where  $R$  and  $R_1$  denote particle radii [21]. In order of their appearance the acceleration terms signify inertia, Hertzian elasticity, viscous dissipation and adhesion. We solved eq. (10) numerically and obtained a critical velocity (fig. 2), which is defined as the largest impact speed

$g_c = \dot{\xi}(t=0)$  for which particles still stick together. It is sensitive to material properties, but agrees with experimental studies for centimetre-sized ice particles [23, 24] in terms of a restitution coefficient. Changing the material used, naturally also  $g_c$  is changed in its numerical values, but nevertheless the functional dependence remains. For masses larger than 100 g (*i.e.* centimetre-sized icy grains) an approximate power law scaling is obtained [22]:

$$g_c(m, \mu) \propto \sqrt{\frac{1+\mu}{\mu m}} = \sqrt{\frac{1}{m_{\text{eff}}}}, \quad (11)$$

with the ratio  $\mu = m_1/m$ . Smaller particles are more likely to stick together than larger ones, whereas larger bodies have to have a very low (mm/s) impact speed to agglomerate. The dependence on  $\mu$  is shown in the right part of fig. 2. With increasing effective mass  $m_{\text{eff}}$  (see [21]), eq. (11) becomes quite accurate but, on the other hand, the gravitational influence gets dominant. The escape velocity

$$v_{\text{esc}} = \sqrt{\frac{2G(m+m_1)}{R+R_1}} \propto m^{1/3} \sqrt{\frac{1+\mu}{1+\mu^{1/3}}}, \quad (12)$$

represents a good approximation for gravitational encounters and is illustrated in grey lines and as a grey shaded area in the left and right part of fig. 2, respectively. The intersections between the grey and the black curves bound the range of validity of eq. (10), *i.e.* in cases of larger  $m$ , gravity in combination with dissipation determines whether the particles do rebound or not. Additionally, we checked the same treatment of the collision dynamics for the elasto-plastic contacts [25] including adhesion which results in a qualitatively similar scaling (Weber number) and domain diagram. For an adhesive elasto-plastic collision one obtains a sticking velocity of  $g_c \propto m^{-5/18}$  (skipping the  $\mu$ -dependence). Equations (10) and (12) cover qualitatively well the transition from domains dominated by adhesion on the one hand and gravity on the other. The sticking domain can be increased to larger  $g_c(m, \mu)$  due to the presence of gas drag forces [26, 27]. It is worth noting that for very low  $\mu$ -values, *i.e.* for very different particle sizes, the critical velocity  $g_c$  rises dramatically owing to adhesion. Since the contact dynamics and  $g_c$  in eq. (11) are governed by the effective mass  $m_{\text{eff}}$ , the actual mass of larger bodies is less important while any significantly smaller particle easily sticks to them ( $\mu \ll 1$ ,  $\mu \gg 1$ ).

*Solution of the coagulation equation.* – We numerically solved eqs. (7)-(9) carefully accounting for the coagulation domain  $\mathcal{D}_{(c)}$  in terms of numerical results of fig. 2. Therefore, gain and loss integrals have been approximated by corresponding sums in which the independent variables, time  $t$  and mass  $m$ , have been discretized. We chose approximately 5000 adaptive time steps within the interval of  $t \in (0, 10^{12})$  s ( $\approx 31710$  y) and 200 mass bins from  $10^{-9}$  kg to  $10^6$  kg. In order to define the particle number density, the total mass and the mass density for the initial state, we assume an Earth mass evenly spread in a circumsolar disk with an semi-opening angle of 3 degrees placed between the orbits of Venus and Mars. The initial distribution reads  $n(t=0) = C_0 m \exp[-C_1 m]$ , where  $C_0$  and  $C_1$  are constants. The character of the obtained solution (see also fig. 3) is insensible to changes of the initial conditions. While collision frequency and growth rate do change, time is scalable in terms of the disk parameters. The numerical code was tested using initial conditions and analytical solutions of the Smoluchowski equation derived by Silk and Takahashi [6] and we found a good agreement. The result of the numerical solution is unsurprisingly coagulation that evolves to higher masses during time. But differently from any growth process predicted by the Smoluchowski equation (sticking at any given contact) general growth went on until reaching a certain size where it almost ceased to continue. This particular size is determined by the ratio of  $g_c$  and

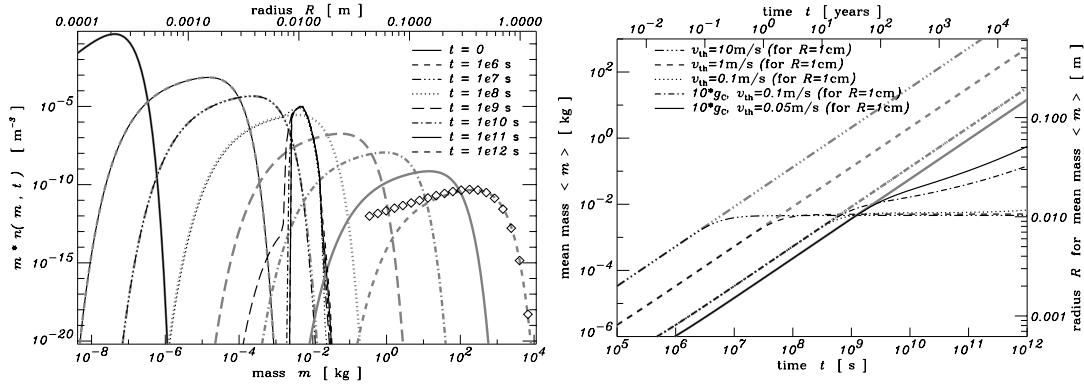


Fig. 3 – Numerical simulations. Left: the time evolution of the mass spectrum  $m n(m)$  in case of  $C = 1$  (grey lines) and  $C(g_c, v_{th})$  according eq. (9) (black lines), where  $v_{th} = 1$  m/s and  $R = 0.01$  m. Any pair of graphs refers to a distinct time step. Snapshots in time are taken for each elapsed order of magnitude, showing a perfect match to a Smoluchowski evolution at the beginning but a drastic difference for later stages. Right: the influence of thermal velocity  $v_{th}$  and threshold velocity  $g_c$  is shown, plotting the mean mass  $\langle m \rangle$  over time. Whenever  $v_{th} \gg g_c$  the growth process is almost stopped, which implies that it takes aeons for larger bodies to form. With ten times higher  $g_c$  (or ten times lower  $v_{th}$ ) growth can be continued at a slower rate beyond the critical size of 1 cm.

$v_{th}$ . Coagulation can still take place, but the growth rate is enormously decreased. In fig. 3 the mass distribution for different time steps is shown (black lines) and compared to results obtained using  $C = 1$  and thus the Smoluchowski equation (grey lines). For the thermal velocity we used  $v_{th} = 1$  m/s for 1 cm particle as given in [28] for the sake of possible astrophysical applications. All simulations are initiated from the same initial distribution. It is clearly visible that up to  $10^7$  s growth evolves in the exact same manner for  $C \rightarrow 1$  and  $C(g_c, v_{th})$ . After that for  $C(g_c, v_{th})$  the creation of larger bodies is prevented while the width of the distribution decreases. Thus, due to restricted coagulation an approximately monodisperse distribution establishes. The diamonds in the left part of fig. 3 represent an approximate asymptotic analytical solution [6] for a power law kernel  $\mathcal{K} \propto m^{\lambda/2} m_1^{\lambda/2}$  ( $\lambda = 1/6$ ), with Dirac's delta-function as the initial condition, and  $C = 1$ . The differences in lower masses arise due to the assumed power law kernel and initial delta-function. Nevertheless, the agreement further validates our numerical code and gives analytic tools to study unrestricted growth in time ( $C \rightarrow 1$ ). For instance, we obtain the rate of growth as  $\langle m \rangle \propto \varrho^{6/5} T^{3/5} t^{6/5}$ , where  $\varrho$  is the disk mass density ( $\varrho = \int m n(m) dm$ ), and  $T$  the granular temperature. The right panel in fig. 3 shows  $\langle m \rangle$  as a function of time for different granular temperatures  $T \propto v_{th}^2$  and  $g_c$ , obtained from simulations with  $C = 1$  and  $C(g_c, v_{th})$ . In the latter case, the growth process ceases at the same critical mass. Increasing  $g_c$  and/or decreasing  $v_{th}$ , growth can again be restored, but at a slower rate. Variations of  $v_{th}$  also shift the onset of laggard growth. As long as  $v_{th} > g_c$  holds, the growth process will be inhibited during its evolution. To provoke a case of perfect coalescence the thermal velocity and thus the granular temperature has to be small enough, implying a dynamically cold ensemble. Since  $g_c$  is a material-dependent quantity, perfect coalescence cannot be obtained for thermal velocities applied here. In case of silicates, ice, or metals-rock composites  $g_c$  remains fairly small.

*Discussion.* – The derived kinetic equations incorporate all possible collisional outcomes. For the sake of brevity, we concentrated on coagulation only, although a common treatment including fragmentation and restitution is a part of ongoing work. Despite knowing that a



Maxwellian velocity distribution and also the separation ansatz of the generalized distribution function is not appropriate for a general discussion, we were able to give an analytical derivation of the otherwise phenomenological Smoluchowski equation. As a result, it is only possible to apply this equation to systems where sticking occurs at any given contact and energy equipartition is true. Furthermore, including a more detailed velocity distribution, as, *e.g.*, an anisotropy of the thermal velocities, does not change the qualitative result of a restricted coagulation. After a certain time an almost monodisperse mass distribution establishes and very slowly develops in favour of larger bodies. These results suggest severe implications for the scenario of planetary growth. An “easy” transition from centimetre-sized particles to planetesimals does not occur for parameters used in this paper. Without other physical effects that generate massive bodies, *i.e.* approximately  $10^3$  kg, no growth on appropriate time scales is possible.

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This work was supported by the Studienstiftung des deutschen Volkes and by the Deutsche Forschungsgemeinschaft (DFG), grant number *Sp 384/12-3*.

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