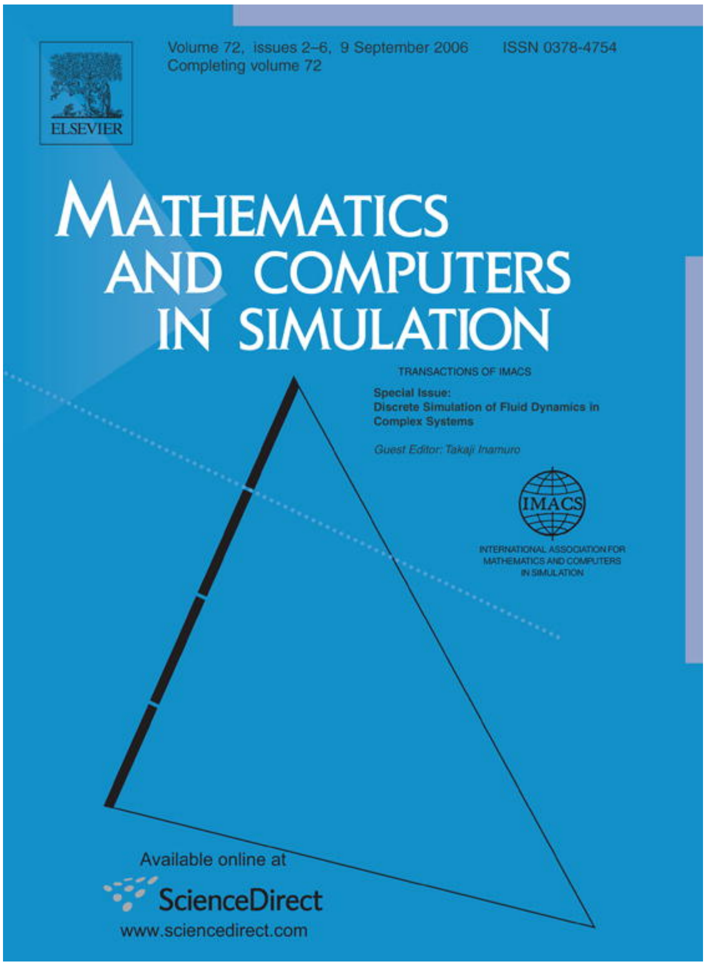
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Dust coagulation in equilibrium molecular gas

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Abstract

We study coagulation of dust particles immersed in equilibrium molecular gas. The dissipative grain interactions are modeled by a constant coefficient of restitution, while for the adhesive interactions the model of Johnson, Kendall and Roberts is applied. We formulate the Boltzmann-type master equation for the mass–velocity distribution function of aggregates and use the Kramers–Moyal expansion for the grain–gas collision integral. We derive coagulation equations and kinetic coefficients for these equations and analyze different regimes of the system evolution.

Keywords: Coagulation; Kinetic coefficient; Equillibrium molecular gas

1. Introduction

There is a vast variety of phenomena, where particles form aggregates upon collisions. The coagulating particles can be molecular-size aggregates (atoms, or clusters) or macroscopic grains, which themselves contain macroscopically large number of atoms. Here we address the latter case, that is, we consider coagulation of grains surrounded by an equilibrium molecular gas. As an example of such phenomena, one can mention atmospheric processes where sedimenting particles, such as water droplets in clouds, dust or soot merge when they meet. The adhesive interactions between particles in these systems are so strong, that each collision results in an aggregation. The particle motion between collisions may be either diffusive, when the density of the surrounding gas is large, or ballistic, when particles move mostly by inertia, slightly decelerated by the dilute surrounding gas. The ballistic aggregation takes place in many astrophysical systems, e.g. in the interstellar dust clouds, dust in the interplanetary space, the faint planetary rings and protoplanetary discs (e.g. [5,6,8]). For the ballistic transport not all collisions are coagulative and the result of an impact depends on the kinetic energy of the relative motion of a colliding pair: if the energy is larger, than a certain threshold, the particles do not form an aggregate and continue to move separately; for very large impact energy the collisions may even become destructive [9]. If the energy, however, is small, the adhesive forces take over and the particles merge. The simplified one-dimensional model of the ballistic aggregation has been first studied by Carnevale, Pomeau and Young (CPY) [4]. The authors assumed that each collision is coagulative and obtained a good agreement with numerical experiments. Later the same model has been studied numerically in [10] with the conclusion that the CPY theory [4]

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is not adequate for dimensions $d \geq 2$. This has been then confirmed in [11], where the authors generalized the model and assumed that the aggregation occurs with some probability, independent on the relative velocity. A more realistic model for ballistic agglomeration has been proposed in [9], where the dissipative particle collisions were explicitly studied. The authors found numerically the impact-velocity-dependent condition for a coagulative collision and derived the Smoluchowski equation for the size distribution of aggregates. They used the approximation of a constant granular temperature T and did not consider its evolution. Moreover, it was assumed that particles move in vacuum, i.e. no interactions of the particles with a surrounding gas were considered.

In the present study we investigate the aggregation kinetics of particles moving in a surrounding gas and analyze the evolution of the particle size distribution, and granular temperature.

2. Particles interactions

2.1. Interaction between grains

For simplicity we assume that all grains have spherical shape (that is, the shape of aggregates relaxes between collisions) and neglect their rotational motion. Since the grains are macroscopic bodies, they always collide dissipatively [3]. The energy loss in the non-aggregative impact of two particles of mass $m_i = m_0 i$ and $m_j = m_0 j$ (m_0 is some minimal mass) and velocities $\vec{v}_i$ and $\vec{v}_j$ is quantified by the coefficient of restitution ε [3]:

$$\vec{v}_i' = \vec{v}_i - \frac{\mu_{ij}(1+\varepsilon)}{m_i} g_{\|}\vec{e}, \qquad \vec{v}_j' = \vec{v}_j + \frac{\mu_{ij}(1+\varepsilon)}{m_j} g_{\|}\vec{e}, \tag{1}$$

where $g_{\|} = (\vec{v}_i - \vec{v}_j)\vec{e}$, the unit vector $\vec{e} \equiv \vec{r}_{ij}/|\vec{r}_{ij}|$ specifies the relative particle position at the collision instant, $\mu_{ij} = m_i m_j/(m_i + m_j)$ the reduced mass and $\vec{v}_i'$ and $\vec{v}_j'$ are the post-collision velocities. Generally the coefficient of restitution depends on the relative velocity [3]. In order to keep the model simple and the analyzed effects accessible, we assume that $\varepsilon =$ constant. It follows from Eq. (1) that the energy loss in a collision is proportional to $(1 - \varepsilon^2)$, that is, it vanishes for $\varepsilon = 1$.

The adhesive forces between particles are described by the Johnson, Kendall and Roberts (JKR) model [7]. It gives the total force between two colliding elastic spheres of radii R_i and R_j and compression $\xi = (R_i + R_j) - |\vec{r}_i - \vec{r}_j|$ in terms of the contact radius a as (see also [2])

$$\xi(a) = \frac{a^2}{R_{\rm eff}} - \sqrt{\frac{8\pi\gamma D a}{3}}, \qquad F(a) = \frac{a^3}{D R_{\rm eff}} - \sqrt{\frac{6\pi\gamma}{D}} a^{3/2}. \tag{2}$$

Here the adhesion coefficient γ characterizes the adhesive energy per unit area, $D = 3(1-\nu^2)/2Y$ describes the mechanical properties of the particles material (ν is the Poisson ratio and Y is the Young modulus) and $R_{\rm eff} = R_i R_j/(R_i + R_j)$. At zero applied load the contact radius a_0 is finite, $a_0^3 = (3/2) D\pi\gamma R_{\rm eff}^2$, while for negative load it decreases until the contact between particles is lost at $a_{\rm sep} = 2^{-2/3} a_0$. Using these values of a_0 and $a_{\rm sep}$ and Eq. (2) we obtain the work requested to separate particles [2],

$$W_{ij} = \int_{a_0}^{a_{\rm sep}} F(a) \frac{d\xi}{da} da = q_0 (\pi^5 \gamma^5 D^2 R_{\rm eff}^4)^{1/3}, \quad q_0 \simeq 1.46. \tag{3}$$

Consider an impact with an initial velocity g. Close to the end of the collision, just before the tensile forces start, the relative velocity is $g' = \varepsilon g$. Then the final velocity g'', at the complete separation, may be found from the conservation of energy, neglecting dissipation during the last part of the collision:

$$\frac{\mu_{ij}(g'')^2}{2} - \frac{\mu_{ij}(g')^2}{2} = W_{ij}. \tag{4}$$

Since coagulation implies $g'' = 0$, Eq. (4) with $g' = \varepsilon g_{\rm st}$ gives the sticking condition

$$g_{\rm st}^2 = \frac{2 W_{ij}}{\varepsilon^2 \mu_{ij}}. \tag{5}$$

Hence, if the relative collision velocity is smaller than $g_{\rm st}$ the particles stick together. Strictly speaking this analysis is valid only for a head-on collision, when the above condition applied to the normal component of the relative velocity $g_{\|}$. The tangential component is expected to be restricted by some other condition. In what follows we, nevertheless, assume the same limitation for the tangential component, which yields the sticking condition, $|\vec{v}_{ij}| < g_{\rm st}$.

2.2. Interactions between gas particles and grains

Since the gas particles are of molecular-size their interactions are elastic. Moreover, we assume that the gas is very dilute and is kept by some mechanism (e.g. by the solar radiation) at thermodynamic equilibrium with constant temperature $T_{\rm g}$. To describe collisions between grains and the gas particles we assume that the particles are smooth (that is, we ignore their rotation) and adopt two collision models: (i) a simplified model of the elastic impact, which conserves the momentum and energy of the colliding particles and (ii) a stochastic collision model, which conserves the momentum, but not energy. Such model describes the exchange of energy between the internal degrees of freedom of a grain and a molecule of the surrounding gas. The gain or loss of energy in such collisions is characterized by a stochastic restitution coefficient $\varepsilon_{\rm g}$ [1]. Hence we apply the collision rule (1), where m_j and μ_{ij} are to be substituted, respectively, by $m_{\rm g}$ and $\mu_{i{\rm g}}$, where $m_{\rm g}$ is the mass of the gas particles, and ε by $\varepsilon_{\rm g}$. The restitution coefficient $\varepsilon_{\rm g}$ describes the loss or gain of the kinetic energy of the relative motion of a colliding pair. This amount of energy, proportional to $\varepsilon_{\rm g}^2 - 1$, is transmitted to (for $\varepsilon_{\rm g} < 1$), or taken (for $\varepsilon_{\rm g} > 1$) from the internal degrees of freedom of a grain. We assume that the *grain material* and the molecular gas are in thermal equilibrium, hence $\langle \varepsilon_{\rm g}^2 \rangle - 1 = 0$.

3. Master equation

The mass–velocity distribution function for the grain particles of the dust–grain mixture $f(m_i, \vec{v}_i, t)$ satisfies the following master equation:

$$\frac{\partial}{\partial t} f(m_i, \vec{v}_i, t) = I_{{\rm g},i}(f_{\rm g}, f_i) + \sum_{j \neq i} I_{ij}^{\rm rest}(f_i, f_j) + \sum_{j \neq i} I_{ij}^{\rm coag}(f_i, f_j), \tag{6}$$

where $I_{{\rm g},i}$, $I_{ij}^{\rm rest}$ and $I_{ij}^{\rm coag}$ denote the collision integrals: $I_{{\rm g},i}$ describes the collisions between the gas particles and grains of mass m_i, while the other two collision integrals $I_{ij}^{\rm rest}(f_i, f_j)$ and $I_{ij}^{\rm coag}$ describe correspondingly the *restitutive* and *coagulative* collisions of grains with mass m_i and m_j. Since the mass ratio for the gas and grain particles is very small, $\Delta_i = m_{\rm g}/m_i \ll 1$, the collision integral $I_{{\rm g},i}$ may be written using the Kramers–Moyal expansion [3]:

$$I_{{\rm g},i}(f_g, f_i) = \frac{\partial}{\partial \vec{v}_i} \left(\gamma_i \vec{v}_i + \bar{\gamma}_i \frac{\partial}{\partial \vec{v}_i} + \cdots \right) f(m_i, \vec{v}_i, t), \tag{7}$$

with constants $\gamma_i = (16/3) n_{\rm g} R_i^2 (2\pi k_{\rm B} T_{\rm g}/m_{\rm g})^{1/2} \Delta_i$ and $\bar{\gamma}_i = (k_{\rm B} T_{\rm g}/m_{\rm g}) \gamma_i$. Here $k_{\rm B}$ is the Boltzmann's constant and $n_{\rm g}$ is the number density of the molecular gas. We also take into account that $\langle \varepsilon_{\rm g}^2 \rangle = 1$, which leads for $\Delta_i \ll 1$ to the same result for both gas–grain impact models. The other two collision integrals read [3,9],

$$I_{ij}^{\rm rest} = \sigma_{ij}^2 \int {\rm d}\vec{v}_j \, {\rm d}\vec{e} |g_{\|}| \Theta(-g_{\|}) \left\{ \varepsilon^{-2} f(m_i, \vec{v}_i'') f(m_j, \vec{v}_j'') \Theta(\varepsilon^{-1} g_{\|}^2 - g_{\rm st}^2) - f(m_i, \vec{v}_i) f(m_j, \vec{v}_j) \Theta(g_{\|}^2 - g_{\rm st}^2) \right\}, \tag{8}$$

$$\begin{aligned} I_{ij}^{\rm coag} = {} & \frac{1}{2} \sigma_{i-j\,j}^2 \int {\rm d}\vec{v}_j \, {\rm d}\vec{e} |g_{\|}| \Theta(-g_{\|}) f(m_{i-j}, \vec{v}_{i-j}) f(m_j, \vec{v}_j) \Theta(g_{\rm st}^2 - g_{\|}^2) \\ & - \sigma_{ij}^2 \int {\rm d}\vec{v}_j \, {\rm d}\vec{e} |g_{\|}| \Theta(-g_{\|}) f(m_i, \vec{v}_i) f(m_j, \vec{v}_j) \Theta(g_{\rm st}^2 - g_{\|}^2), \end{aligned} \tag{9}$$

where $\Theta(x)$ is the unit step function, $\sigma_{ij} = R_i + R_j$ and $\vec{v}_i''$, $\vec{v}_j''$ are initial velocities of the *inverse* collision, that is of the collision that ends up with the velocities $\vec{v}_i$, $\vec{v}_j$ [3].

4. Coagulation kinetics

Let $n_i = \int d\vec{v}_i f(m_i, \vec{v}_i)$ be the number density of granular particles of mass m_i, and $n = \sum_i n_i$ the total number density. We also introduce partial *granular* temperature T_i and the total *granular* temperature T as,

$$\frac{3}{2} n_i T_i = \frac{1}{2} \int d\vec{v}_i f(m_i, \vec{v}_i) m_i \vec{v}_i^2, \quad nT = \sum_i n_i T_i. \tag{10}$$

Let the partial and total granular temperatures are equal, and the distribution functions are Maxwellian: $f(m_i, \vec{v}_i, t) = n_i (m_i/2\pi T)^{3/2} \exp(-m_i v_i^2/2T)$. Then multiplying Eq. (6) with 1 and with $m_i v_i^2/2$ and integrating over $\vec{v}_i$ we obtain,

$$\frac{\partial}{\partial t} n_k = \frac{1}{2} \sum_{i+j=k} C_{ij} n_i n_j - n_k \sum_j C_{ij} n_j \tag{11}$$

$$\frac{\partial}{\partial t} nT = 3(k_B T_g - T) \sum_i n_i \gamma_i - \sum_j D_{ij} n_i n_j, \tag{12}$$

with the kinetic coefficients which depend on $w_{ij} \equiv \mu_{ij} g_{st}^2/2T = W_{ij}/\varepsilon^2 T$:

$$\begin{aligned} C_{ij} &= 2\sigma_{ij}^2 \left(\frac{2T\pi}{\mu_{ij}}\right)^{1/2} (1 - (1 + w_{ij}) e^{-w_{ij}}) \\ D_{ij} &= \left(\frac{\sigma_{ij}^2}{3}\right) \left(\frac{2T^3\pi}{\mu_{ij}}\right)^{1/2} e^{-w_{ij}} [4e^{w_{ij}} - (1 + \varepsilon^2)(2 + 2w_{ij} + w_{ij}^2)] \end{aligned} \tag{13}$$

The Smoluchowski-like Eqs. (11)–(13) are the main results of our study. The general analysis of this infinite set of nonlinear equations is rather complicated; some limiting cases, however, may be studied analytically. There are two basic quantities which control the system evolution, $W_{ij}/k_B T_g$ and $W_{ij}/T(0)$. Referring for details to the forthcoming publications, we briefly sketch the results of the analysis:

- In the limit $W_{ij}/k_B T_g \ll 1$, $W_{ij}/T(0) \ll 1$ for all i and j, the coagulation is suppressed and n_i and n are kept constant. In this regime the granular temperature relaxes (from above or from below) to the stationary temperature, T^*, which is the solution of the equation: $T^* = 3k_B T_g \gamma/(3\gamma + \zeta(T^*))$. Here $\gamma = \sum_i \gamma_i n_i/n$ and $\zeta(T) = \sum_{ij} \sigma_{ij}^2 (8\pi T/9\mu_{ij})^{1/2}(1 - \varepsilon^2) n_i n_j/n$. Note that in the final state the granular and gas temperatures are *different*.
- In the limit $W_{ij}/k_B T_g \ll 1$, $W_{ij}/T(0) \gg 1$ all collisions between grains are initially coagulative and the granular temperature and the mean mass of the aggregates $\langle m \rangle$ grow with time as $T \sim t^{4/3}$ and $\langle m \rangle \sim t^2$. This implies the decay of the average ratio $\langle W_{ij} \rangle / T \sim 1/t^{4/9}$ which controls the coagulation. Hence the coagulation will gradually cease and the system will arrive to the non-coagulative steady state with $T = T^*$.
- In the limit $W_{ij}/k_B T_g \gg 1$, $W_{ij}/T(0) \gg 1$ the collisions are always coagulative. As $t \to \infty$ the coagulation continues and granular temperature tends asymptotically to the gas temperature, $T \to k_B T_g$.
- In the limit $W_{ij}/k_B T_g \gg 1$, $W_{ij}/T(0) \ll 1$ the granular temperature initially decays without coagulation, which starts however at later stage. Finally, the system evolves to the state described previously: The granular temperature tends to $k_B T_g$ and particles continue to coagulate at each collision.

In the present study we used a simplified collision model, that is, we developed the aggregation condition for a head-on collision, which restricts the normal component of the impact velocity and applied the same restriction for the tangential component. This approach enormously facilitates the analysis, still reflecting the basic physics of aggregative collisions. In particular, the main conclusions itemized above, remain qualitatively valid.

5. Conclusion

We analyze coagulation of dust particles immersed in the surrounding molecular gas, which is kept at constant temperature. The dissipative collision between grains are described by a constant coefficient of restitution. For the adhesive particle interactions we adopt the Johnson, Kendall and Roberts model. Starting with the general master equation of Boltzmann type, we derived coagulation equations and kinetic coefficients of these equations. We analyze several regimes of the system evolution and show that depending on the gas temperature, granular temperature, properties of the grain material and the gas parameters, two qualitatively different final states of the system are possible—the non-stationary coagulating state with the granular temperature tending to the gas temperature and the stationary non-coagulating state where the gas and granular temperatures are different.

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