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The role of adhesion for ensembles of mesoscopic particles

Y. Baibolatov · F. Spahn

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Abstract We present a toy-model for an ensemble of adhering mesoscopic constituents in order to estimate the effect of the granular temperature on the sizes of embedded aggregates. The major goal is to illustrate the relation between the mean aggregate size and the granular temperature in dense planetary rings. For sake of simplicity we describe the collective behavior of the ensemble by means of equilibrium statistical mechanics, motivated by the stationary temperature established by the balance between a Kepler-shear driven viscous heating and inelastic cooling in these cosmic granular disks. The ensemble consists of N' equal constituents which can form cluster(s) or move like a gas—or both phases may coexist—depending on the (granular) temperature of the system. We assume the binding energy levels of a cluster $E_c = -N_c\gamma a$ to be determined by a certain contact number N_c, given by the configuration of N constituents of the aggregate (energy per contact: $-\gamma a$). By applying canonical and grand-canonical ensembles, we show that the granular temperature T of a gas of constituents (their mean kinetic energy) controls the size distribution of the aggregates. They are the smaller the higher the granular temperature T is. A mere gas of single constituents is sustained for $T \gg \gamma a$. In the case of large clusters (low temperatures $T \ll \gamma a$) the size distribution becomes a Poissonian.

Keywords Adhesion · Statistical mechanics · Planetary rings

Y. Baibolatov (✉) · F. Spahn
Department of Physics and Astronomy, University of Potsdam, Karl-Liebknecht-Str. 24/25, 14476 Potsdam, Germany
e-mail: yernur@agnld.uni-potsdam.de

1 Introduction

Granular matter does come across not only in our daily life, e.g. in a sugar bowl, breakfast cereals, kids sandbox, but also accounts for many interesting and spectacular natural phenomena. Terrestrial examples are avalanches, migrating dunes, sand ripples driven by turbulent wind action, ice floefields in polar oceans, and last but not least, granular gases in the universe comprising the base for the formation of celestial bodies like planets, satellites, comets and asteroids.

Dry granular gases are characterized by dissipative interactions of grains whereas usually attractive forces are neglected. Here we consider attractive-adhesive contacts [1] between the constituents of the granular gas in order to quantify their influence on the cluster formation and the establishment of a steady aggregate size-distribution in driven systems.

In this context the dynamics of dense planetary rings is our major concern. These disks are dynamically dominated by a balance between viscous (collisional) heating, driven by the Kepler-shear, counteracted by granular collisional cooling so that a stationary granular temperature may establish [2]. Interestingly, the Cassini experiments have uncovered that this balance is violated in perturbed regions of planetary rings resulting in an enhanced temperature and decreased particle size accentuating optically these ring-areas [3].

In order to interpret these exciting observations we aim at an estimate of the size distribution of aggregates as a function of the granular temperature of the surrounding gas of monodisperse constituents for a given adhesive energy (per area) at contact γ. This, in turn, means that the full non-linear dynamics [4] between constituents at contact is neglected, and that only adhering attractive bonds are taken into account.

Of course, these are quite radical simplifications because in reality the formation of a contact with a subsequent

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breaking is irreversible and dissipates energy together with viscoelastic deformations of the constituents material during particle contacts [5]. We account for this dissipation by assuming that all motions (oscillations) of the constituents forming the aggregate are quickly dissipated—respective radiated away (partitioned by the restitution coefficient r), so that one is left with certain levels of configurational adhesive binding energy.

Next we claim that the surrounding gas of constituents with a stationary granular temperature is in a "quasi"-equilibrium with one embedded aggregate (consisting of N constituents) and its configurational energetic states, determined by the contact configurations. Their spectrum of binding levels is related to configurational (entropic) temperatures for the surrounding gas to interact with (like in the Ising model [6]).

With this prerequisites we state that statistical mechanics is able to characterize the collective behavior of the ensemble including the aggregate. Again, we emphasize that such a treatment is in principle not valid for dissipative non-equilibrium systems (inelastic collisions, adhesive hysteresis). Contrarywise, the stationary state in planetary rings—where dissipated energy is steadily replenished by the Keplerian shear motion provides arguments in favor of steady state velocity—and size distributions and equilibrium modeling of dissipative systems [7,8]. This reasoning is, for instance, further supported by the usage of free energy functionals in other driven stationary granular systems to identify a "cluster" transition of a granular gas near a van der Waals like critical point [9].

The paper is organized as follows: In Sects. 2 and 3 we quantify the role of adhesion for the granular ensemble by using means of equilibrium statistical mechanics—the canonical and grand-canonical ensembles. And in Sect. 4, we discuss results, methods and assumptions obtained and used here. Further we apply our findings to interpret observations of Saturn's rings obtained by the Cassini spacecraft and give some conclusions for future work.

2 The model

Johnson et al. provided expressions for the adhesive force $F_A \propto \pi\gamma s_{\text{eff}}$ (effective grain radius: $s_{\text{eff}} = s_1 s_2/(s_1 + s_2)$) by means of minimizing the total (free) energy during the contact [1]. In subsequent work [4,5,10] we implemented this surface force in the dynamics of the contact of two colliding granules, where apart from the adhesive force F_A, elastic stresses and viscous dissipative forces determine the dynamics.

Instead of analyzing the rather involved equation of the contact dynamics [4,5] in molecular dynamics simulations, here we focus on the effect of an adhesive attraction, characterized by the contact energy $\varepsilon_c = -\gamma a$, with the specific (per area) adhesive energy γ and the contact area a. In principle, we consider a situation of the aggregate where dissipation has already decayed all vibrations of constituents so that we are left with the binding energy of adhesive contacts ε_c comprising certain energetic levels respective to corresponding entropic temperatures. Next, we argue that statistical mechanics is suitable to gain information and trends about the affinity of the ensemble to form clusters as a function of the (in driven systems constant) granular temperature [8].

The system under inspection is an "isothermal" gas of constituents with the above mentioned properties in thermal contact with a single embedded aggregate. After all, the probability distribution for the aggregate size derived by means of statistical mechanics is then equivalent to the size distribution of a system of non-interacting aggregates embedded in a gas of adhesive monomers.

3 Statistical mechanics

In our model we consider identical adhesive spheres, which can stick to each other and form aggregates. If two particles are sticking and forming a bond of an area a, the energy of the bond becomes

$$\varepsilon_c = -\gamma a, \tag{1}$$

whereas oscillations inside the aggregate are considered to have decayed (dissipated energy is radiated in space). In other words all the bonds are assumed to be static, and a certain configuration of adhesive contacts, carrying the energy (1), constitutes the binding energy level

$$E_c = -N_c \gamma a, \tag{2}$$

which may adjust according to the temperature of the surrounding granular gas (averaged kinetic energy per constituent). The number of contacts of the aggregate in a certain thermal/entropic configuration is N_c.

Aggregates with a given number of constituents can have different numbers of contacts among the constituents due to differences in the configuration. Considering all possible configurations for a given number of constituents N, we obtain a discrete spectrum of energetic levels which the aggregate can take.

The configuration with minimal number of contacts (maximal binding energy level) is obviously a linear chain of particles, illustrated in Fig. 1. This configuration has $N_c = N - 1$ bonds corresponding to highest energy level labeled with the index $i = 0$. Now, from this linear chain one may construct the next configuration with $N_c = N$ contacts, i.e. adding one more contact (Fig. 1) and assigning an index $i = 1$. Continuing this operation, we end up with the configuration which has the maximal possible number of contacts for the given number of constituents. This configuration corresponds to the

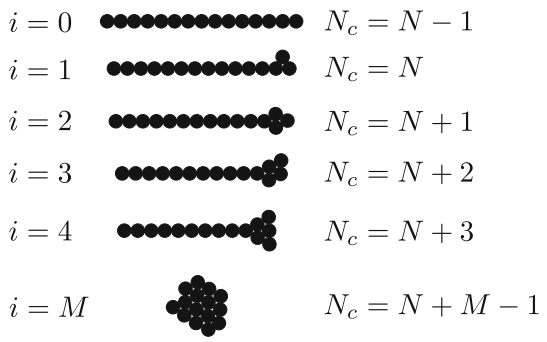

Fig. 1 The sketch of possible configurations of an aggregate with N constituents. We can see, that the chain configuration has minimum number of contacts and denoted as $i = 0$ corresponding to the maximum energy. The configuration with maximum number of contacts is denoted as $i = M$, setting the ground state of minimal aggregate energy. On the *right column* the numbers of contacts are shown for given configuration

lowest possible binding level—called ground state—characterized with index $i = M$.

All possible configurations comprise a spectrum of energetic levels

$$\left\{E_c^{(i)}\right\} = \left\{E_c^{(0)}, E_c^{(1)}, E_c^{(2)}, \ldots, E_c^{(M)}\right\}. \tag{3}$$

Each energetic level $E_c^{(i)}$ is ν_i-times degenerated so that the canonical partition function of our model reads:

$$Z_N = \frac{1}{N!}\sum_{i=0}^{M} \nu_i e^{-\beta E_c^{(i)}}, \tag{4}$$

with the parameter $\beta = 1/T$. Here indistinguishable constituents are assumed explaining the pre-factor $1/N!$ and simplifying the further analytical treatment.

It seems plausible to assume minimal values $\nu_0 = \nu_M = 1$ and increasing degeneracy functions between these (for an aggregate) limiting levels as for instance $\nu_i = M!/[i!(M-i)!]$, which also permits to derive a closed solution of the sum

$$Z_N = \frac{1}{N!}\sum_{i=0}^{M} \binom{M}{i} e^{-\beta E_c^{(i)}}, \tag{5}$$

or

$$Z_N = \frac{1}{N!}\sum_{i=0}^{M} \binom{M}{i} e^{(N+i-1)\beta\gamma a} = \frac{x^{N-1}}{N!}\sum_{i=0}^{M} \binom{M}{i} x^i, \tag{6}$$

with $x = \exp(\beta\gamma a) \geq 1$. For very large values of N (and correspondingly M) we may write for the partition function:

$$Z_N = \frac{x^{N-1}}{N!}\cdot(1+x)^M = \frac{x^{N+M-1}}{N!}\cdot\left(1+\frac{1}{x}\right)^M, \tag{7}$$

to give

$$Z_N = \frac{e^{N_c^{(M)}\beta\gamma a}}{N!}\cdot\left(1+e^{-\beta\gamma a}\right)^M, \tag{8}$$

where $N_c^{(M)} = N + M - 1$ is the number of contacts for the ground state configuration.

3.1 Thermodynamic functions

Knowing the partition function (8), we can derive thermodynamic functions, as the free energy, entropy and the internal energy of the aggregate:

$$\begin{aligned}
\ln Z_N &= N_c^{(M)}\beta\gamma a - \ln N! + M\ln\left(1+e^{-\beta\gamma a}\right),\\
F(T) &= -T\ln Z_N\\
&= T\ln N! - N_c^{(M)}\gamma a - MT\ln\left(1+e^{-\beta\gamma a}\right),
\end{aligned} \tag{9}$$

$$\begin{aligned}
S(T) &= -\frac{\partial F(T)}{\partial T}\\
&= -\ln N! + M\ln\left(1+e^{-\beta\gamma a}\right) + \frac{M\gamma a}{T}\frac{1}{1+e^{\beta\gamma a}},
\end{aligned} \tag{10}$$

$$U(T) = F(T) + TS(T) = -\left(N_c^{(M)} - \frac{M}{1+e^{\beta\gamma a}}\right)\gamma a,$$

or

$$U(T) = -\left(N_c^{(M)} - Mf(T)\right)\gamma a. \tag{11}$$

The introduced function $f(T) = \left(1+e^{\beta\gamma a}\right)^{-1}$ vanishes at $T \to 0$ and is monotonically increasing with T. Now we can see that at very low temperatures, the aggregate is in its ground state with the maximum number of contacts $N_c^{(M)}$ and with the minimum of energy $U(T) = -N_c^{(M)}\gamma a$. For $T \to \infty$, $f(T) = 1/2$, and the maximum value of energy is $U(T) = -\left(N_c^{(M)} - M/2\right)\gamma a$.

3.2 Grand canonical ensemble

Next, the aggregate is put in contact with its granular gas, where the impacting particles can stick to the aggregate due to adhesion or can erode the aggregate, changing the number of constituents of the aggregate. Further, impacting (even restituting) gas-constituents can also change the aggregate-configuration, and with it, the binding level $E_c^{(i)}$.

Then, consider that our aggregate (and its energetic binding energy levels) is in a dynamical "quasi"-equilibrium with the granular gas and that the grand canonical ensemble can be applied with the grand partition function:

$$Q(\mu, T) = \sum_{N=0}^{\infty} e^{\beta\mu N}\cdot Z_N(T), \tag{12}$$

where μ is the chemical potential. With Eq. (8) one may write

$$Q(\mu, T) = \sum_{N=0}^{\infty} e^{\beta\mu N} \cdot \frac{e^{N_c^{(M)}\beta\gamma a}}{N!} \cdot \left(1 + e^{-\beta\gamma a}\right)^M . \qquad (13)$$

In order to calculate this sum, we have to make certain assumptions on the maximal number of contacts $N_c^{(M)}$. In the two dimensional case (sticky disks) the maximal number of contacts is calculated as

$$N_c^{(M)} = \left[3N - \sqrt{12N - 3}\right], \qquad (14)$$

where $[x]$ denotes the greatest integer less than or equal to x [11]. For very large N, we can approximate Eq. (14) as $N_c^{(M)} \approx 3N - 2\sqrt{3N} = \lambda_v^{(2)} N - \lambda_s^{(2)}\sqrt{N}$. Here we split the maximal number of contacts into the volume part $\lambda_v^{(2)} N$ and the surface part $\lambda_s^{(2)}\sqrt{N}$. The coefficients $\lambda_v^{(2)} = 3$ and $\lambda_s^{(2)} = 2\sqrt{3}$ have the meaning of the mean number of contacts per particle, correspondingly inside the aggregate and on its surface. The superindex $^{(2)}$ denotes the two dimensional case.

This approximate relation between the maximum number of contacts and the total number of constituents is quite easy to show. In the two dimensional case, the volume of the aggregate $V^{(2)}$, e.g. the area, equals $V^{(2)} = \pi\tilde{R}^2$, and the surface, e.g. the length, equals $S^{(2)} = 2\pi\tilde{R}$. Here, $\tilde{R}$ is the radius of the aggregate, where we have assumed that in the configuration with maximum number of contacts, the aggregate is circularly (spherically) shaped [11]. Now, we can write $S^{(2)} = 2\sqrt{\pi V^{(2)}}$. On the other hand, the total volume of the aggregate equals $V^{(2)} = N \cdot v_0$, where v_0 is the volume (the area) of a single constituent (disk). And the surface is $S^{(2)} = 2\sqrt{\pi v_0} \cdot \sqrt{N} = s_0 \cdot \sqrt{N}$, where s_0 is the surface, e.g. the length, of a single constituent. Since all the constituents are equally shaped and sized, it is plausible to expect that on average all of them have the same number of contacts. Hence, the number of contacts in the bulk part of the aggregate can be written as $N_c^{(v)} = \lambda_v^{(2)} V/v_0 = \lambda_v^{(2)} N$.

For the number of contacts on the surface of the aggregate one may write $N_c^{(s)} = \lambda_s^{(2)} S/s_0 = \lambda_s^{(2)}\sqrt{N}$, with $\lambda_v^{(2)}$ and $\lambda_s^{(2)}$ being the proportionality constants. Finally we have:

$$N_c^{(M)} = \lambda_v^{(2)} N - \lambda_s^{(2)}\sqrt{N}\,, \qquad (15)$$

affirming Eq.(14).

The subtraction arises from the fact, that $\lambda_v^{(2)} N$ slightly overestimates the number of contacts, because the constituents on the surface have less contacts than the ones inside the aggregate.

The three dimensional case could be derived using the same arguments giving:

$$N_c^{(M)} = \lambda_v^{(3)} N - \lambda_s^{(3)} N^{2/3}. \qquad (16)$$

However, for the following analysis, we will assume the total number of contacts to be $N_c^{(M)} = \lambda_v^{(3)} N\left(1 - O(N^{-1/3})\right) \approx \lambda_v N$ and $M \approx (\lambda_v - 1)N + 1$, i.e. an approximation for large aggregates. This allows us to solve the grand partition function analytically and Eq. (13) can be written as

$$Q(\mu, T) = \sum_{N=0}^{\infty} \frac{e^{N\beta(\mu+\lambda_v\gamma a)}}{N!} \cdot \left(1 + e^{-\beta\gamma a}\right)^{(\lambda_v-1)N+1}, \qquad (17)$$

or

$$Q(\mu, T) = \psi(T) \sum_{N=0}^{\infty} \frac{\left(\psi(T)^{\lambda_v - 1} \cdot e^{\beta(\mu+\lambda_v\gamma a)}\right)^N}{N!}, \qquad (18)$$

where $\psi(T) = 1 + e^{-\beta\gamma a}$. Equation (18) constitutes the Taylor expansion of an exponential function which permits to write

$$Q(\mu, T) = \psi(T) \cdot \exp\left(\psi(T)^{\lambda_v - 1} \cdot e^{\beta(\mu+\lambda_v\gamma a)}\right). \qquad (19)$$

Now, we can obtain all the thermodynamic functions for a system with variable number of particles.

First, we will obtain the average number of particles of the aggregate

$$\bar{N} = \frac{1}{\beta}\left(\frac{\partial}{\partial\mu} \ln Q(\mu, T)\right)_\beta ,$$

which gives us

$$\bar{N} = \psi(T)^{\lambda_v - 1} \cdot \exp\left(\frac{\mu + \lambda_v\gamma a}{T}\right). \qquad (20)$$

The number $\bar{N} \to \infty$ diverges for $T \to 0$ in the thermodynamic limit (like at critical points for phase transitions), whereas the mean sizes decay exponentially with growing temperature T.

The internal energy can be written as:

$$U(\mu, T) = -\left(\frac{\partial}{\partial\beta} \ln Q(\mu, T)\right)_\mu + \mu\bar{N}, \qquad (21)$$

and using (19, 20) we obtain

$$U(\mu, T) = -\lambda_v\bar{N} \cdot \gamma a - \frac{(\lambda_v - 1)\bar{N} + 1}{\psi(T)} \cdot \frac{d\psi}{d\beta}. \qquad (22)$$

Here $\lambda_v\bar{N} = \bar{N}_c$ is the average number of contacts for given T. From the above estimates, we can write $\bar{M} = (\lambda_v - 1)\bar{N} + 1$, the index of the average (or the most probable) configuration. Finally, the expression for the internal energy reads:

$$U(\mu, T) = -\left(\bar{N}_c - \bar{M} f(T)\right)\gamma a. \qquad (23)$$

For very low temperatures $T \to 0$, the function $f(T)$ vanishes and we arrive again at the ground state $U(\mu, T) = -\bar{N}_c\gamma a \approx -\lambda_v\bar{N}\gamma a$.

3.3 Aggregate size distribution

Equation (12) can be written in the following form

$$Q(\mu, T) = \sum_{N=0}^{\infty} Q_N(\mu, T), \tag{24}$$

where

$$Q_N(\mu, T) = e^{\beta\mu N} \cdot Z_N(T) = \psi(T) \cdot \frac{\bar{N}^N}{N!}. \tag{25}$$

With this, the probability of the ensemble to have N particles is

$$p(N, \mu, T) = \frac{Q_N(\mu, T)}{Q(\mu, T)}, \quad \sum_N p(N, \mu, T) = 1. \tag{26}$$

Note, that the ensemble is already averaged over all possible energetic states for given N, hence we use Eq. (19) and arrive at

$$p(N, \bar{N}) = \frac{\bar{N}^N}{N!} \cdot e^{-\bar{N}}, \tag{27}$$

which is the Poisson distribution with average $\bar{N}$, valid for larger (colder) aggregates. Figure 2 shows the above distribution (27) and is obtained using the scheme: we fix the total number of constituents N_{total} and the parameters $\mu + \lambda_v \gamma a = 1$. Then we change the temperature T, from the values $T \ll \mu + \lambda_v \gamma a$ up to $T \gg \mu + \lambda_v \gamma a$. For each value of T, we construct a curve from (27) varying N in range $[1 \div N_{\text{total}}]$ and normalizing it with (26). We can see from Fig. 2 that for high values of $T \gg \mu + \lambda_v \gamma a$, the aggregates with small numbers of constituents are more probable (red curve). In contrary, for low temperatures $T \ll \mu + \lambda_v \gamma a$, all constituents try to form a single large aggregate (black curve).

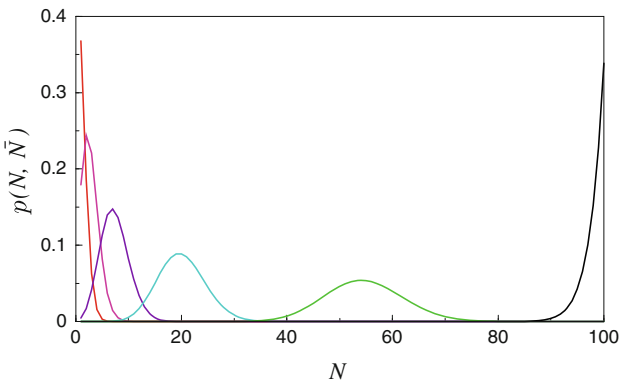

Fig. 2 Aggregates size distribution due to Eq. (27). Different *lines* correspond to different temperatures (T is decreasing from *left* to *right*). The total number of particles $N_{\text{total}} = 100$ is fixed. For the very high temperatures ($T \gg \mu + \lambda_v \gamma a$) very small sized aggregates are more probable to appear (*left side peaks*), while for the low temperatures $T \ll \mu + \lambda_v \gamma a$, the probable size of the aggregate is increasing (*right side peaks*). The intermediate peaks are obtained for $T \sim \mu + \lambda_v \gamma a$. The parameters are taken to be $\mu + \lambda_v \gamma a = 1$

4 Discussion

In this work we have derived a simplified conservative model of an adhering granular gas (of stationary temperature) hosting an aggregate by using methods of equilibrium statistical mechanics: the *canonical* and *grand-canonical* ensemble. We have considered the adhesive contact energy $\varepsilon_c = -\gamma a$ to be the crucial interaction giving rise to available (configurational) energy levels of the aggregate embedded in the model gas. Although being aware of the simplifications used, our "toy-model" clearly accentuates the role of attractive contact forces concerning cluster formation in dependence of the granular temperature in a driven system. The latter can be considered to be constant when cluster formation in shear driven cosmic granular gases—like planetary rings or pre-planetary disks, the nursery of planets—is considered. In similar cases, where dissipated kinetic motion is steadily replenished by outer driving, equilibrium methods have already used successfully [8] supporting the treatment applied here.

The internal energy $U(\mu, \gamma, T)$, the mean particle number (mass, size) $\bar{N}(\mu, \gamma, T)$ and the cluster size-distribution $p(N, \mu, \gamma, T)$ have been found as functions of the specific contact energy γ, the chemical potential μ (related to the contact surface and energy), and the granular temperature T. The temperature T is related to the specific adhesive (potential) binding energy $\varepsilon_c^{(i)} \propto N_c^{(i)}/N$ per constituent, and like in the Ising model, for increasing $T > \gamma a$ entropic contributions more and more dominate energy levels (temperature) of the system. The thermal contact of the granular gas of a given granular temperature T allows the aggregate's energy levels $E_c^{(i)}$ (entropic temperature) and its size ($\bar{N}$) to adjust accordingly.

As a result of the analyses of the corresponding partition sums an increasing mean aggregate size, $\bar{N}$, has been obtained for falling T (see Eq. (20)). This appears to be quite plausible, because entropic contributions become the more suppressed the lower the temperature T is.

Interestingly, these results naturally explain what has been observed in perturbed regions of Saturn's dense rings with the Cassini-cameras [3]. In dynamically excited and perturbed ring-areas an enhancement of the ring-brightness has been measured which is clearly related to a considerable fraction of smaller ring-aggregate sizes. Our analyses show that the enhanced granular temperature in perturbed regions serves as a natural explanation for these Cassini findings provided that Saturn's ring-particles are adhesively bond aggregates.

Using statistical mechanics methods we have demonstrated that our simple "toy" model explains the ability and affinity of the adhering constituents to promote clusters—a process which is driven by the (granular) temperature of the system/surrounding gas. Although a proof to apply equilibrium methods is still needed, the steady

driving of the granular systems of our interest (planetary rings) by the Kepler-shear—resulting in a constant granular temperature—argues in favor of such methods, especially when aiming rather at estimates.

In continuation of the present model, lattice gas approaches could be studied using adhering contacts. Nevertheless, in more advanced models, dissipative effects have either to be addressed by molecular dynamics simulations or by applying a kinetic approach [4].

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