

VIOLETION OF MOLECULAR CHAOS IN DISSIPATIVE GASES

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Numerical simulations of a dissipative hard sphere gas reveal a dependence of the cooling rate on correlation of the particle velocities due to inelastic collisions. We propose a coefficient which characterizes the velocity correlations in the two-particle velocity distribution function and express the temperature decay rate in terms of this coefficient. The analytical results are compared with numerics.

Keywords: kinetic gas theory, many-particle systems, dissipative gases, granular gases, velocity correlations

1. Introduction

The Boltzmann equation for the velocity distribution $f(\vec{v}, t)$ of particles of a gas has been derived with the use of the *molecular chaos* assumption, which implies that all correlations between the velocities of particles may be neglected¹. This equation was also a starting point to build up a kinetic theory of dissipative gases where the particles collide inelastically (for an overview see, e.g.²). Whereas the molecular chaos assumption is justified for dilute molecular gases, there are two reasons for its violation for the case of moderately dense dissipative gases: First the positions of the particles are not independent due to excluded volume effects which gives rise to the Enskog factor.³ A second source of correlations is the dissipative nature of particle interaction. Since dissipation concerns only the normal component of the relative velocity of colliding particles, the angle between their velocity vectors after the collision is always smaller than the angle just before the collision. The latter effect leads to aligning of the trajectories of adjacent particles which contradicts the molecular chaos assumption. Obviously this effect increases with increasing inelasticity, expressed by the coefficient of restitution ϵ which relates the normal component of the relative particle velocity after and before a collision. This expectation has been confirmed by studies of randomly driven Granular Gases⁴, where it was shown that the molecular chaos assumption becomes less accurate with increasing degree of particle inelasticity. Instead, the velocities of the colliding pair are noticeably correlated; this effect depends on the restitution coefficient and on the

gas density.⁵ On the macroscopic (hydrodynamic) scale these velocity correlations cause vortex structures in the velocity field of a spatially homogeneous dissipative gas.⁶

In spite that obviously the microscopic correlations of the velocities of adjacent particles play an important role for the kinetics of dissipative gases, the particular form of the multi-particle velocity distribution function, which describes these correlation is not known. It is the aim of this paper to present the first (to our knowledge) attempt to construct the two-particle correlation function of a simple dissipative gas.

2. Boltzmann-Enskog equation for hard spheres

In kinetic gas theory the evolution of the velocity distribution function $f(v, t)$ is governed by the Boltzmann equation¹

$$\frac{\partial}{\partial t} f(\vec{v}_1, t) = I(f, f) \quad (1)$$

with the collision integral

$$I(f, f) \equiv \sigma^2 \int d\vec{v}_2 \int d\vec{e} \Theta(-\vec{v}_{12} \cdot \vec{e}) |\vec{v}_{12} \cdot \vec{e}| [\chi f(\vec{v}'_1, t) f(\vec{v}'_2, t) - f(\vec{v}_1, t) f(\vec{v}_2, t)] . \quad (2)$$

The velocities \vec{v}_1 and \vec{v}_2 are the pre-collision velocities of the *direct collision*, and the unit vector \vec{e} points from the center of particle 2 with velocity \vec{v}_2 to the center of particle 1 with velocity \vec{v}_1 . Here $\vec{v}_{12} \equiv \vec{v}_1 - \vec{v}_2$, σ is the particle diameter and $\Theta(x)$ is the Heaviside step-function. The velocities \vec{v}'_1, \vec{v}'_2 correspond to pre-collision velocities of the *inverse collision*, which ends up after the collision with the velocities \vec{v}_1 and \vec{v}_2 .

For a gas of inelastically colliding particles the function

$$\chi \equiv \frac{|\vec{g}''|}{|\vec{g}|} \frac{\mathcal{D}(\vec{v}'_1, \vec{v}'_2)}{\mathcal{D}(\vec{v}_1, \vec{v}_2)} \quad (3)$$

accounts for the Jacobian of the transition from the pre-collision velocities \vec{v}'_1, \vec{v}'_2 of the inverse collision to those of the direct collision \vec{v}_1, \vec{v}_2 and for the ratio of the lengths of the collision cylinders of the inverse and the direct collision. These are proportional to the normal components of the relative velocities, i.e. $\vec{g} \equiv (\vec{v}_{12} \cdot \vec{e}) \vec{e}$ for the direct collision, with the analogous expression for the inverse collision. The ratio

$$\epsilon \equiv |\vec{g}| / |\vec{g}''| \quad (4)$$

defines the coefficient of restitution ϵ which describes the inelasticity of the particles. Using this coefficient the velocities of two colliding particles before and after a collision can be related. In particular, these relations read for the inverse collision

$$\vec{v}'_1 = \vec{v}_1 - \frac{1 + \epsilon}{2\epsilon} (\vec{v}_{12} \cdot \vec{e}) \vec{e} \quad (5)$$

$$\vec{v}'_2 = \vec{v}_2 + \frac{1 + \epsilon}{2\epsilon} (\vec{v}_{12} \cdot \vec{e}) \vec{e}. \quad (6)$$

The coefficient of restitution is the central quantity in the theory of granular gases. Experiments⁷ as well as theory^{8,9,10} show clearly that the coefficient of restitution is not a constant but depends on the impact velocity. Nevertheless, for the sake of simplicity of calculation it is frequently assumed that this coefficient is a material constant. With the assumption $\epsilon = \text{const.}$ from (3) we find $\chi = \epsilon^{-2}$.

3. Correlations in dissipative gases

The most important precondition for the derivation of (1,2) is the assumption of molecular chaos (Stoßzahlansatz) which expresses the two-particle distribution function as a product of the one-particle distribution functions

$$f_2(\vec{v}_1, \vec{v}_2, t) = f(\vec{v}_1, t) f(\vec{v}_2, t) \quad (7)$$

which is identical with the assumption of vanishing correlations between successive collisions of each particle. For gases of particles of finite sizes this assumption is violated due to finite volume effects (i.e. two particles can not occupy the same volume). This gives rise to the Enskog factor¹¹ $g_2(\sigma)$

$$f_2(\vec{v}_1, \vec{v}_2) \approx g_2(\sigma) f(\vec{v}_1, t) f(\vec{v}_2, t) \quad (8)$$

which accounts for the increasing collision frequency in such gases. This quantity equals the contact value of the pair correlation function $g_2(\sigma)$. For systems of hard discs it reads:¹²

$$g_2(\sigma) = \frac{1 - \frac{7}{16}\eta}{(1 - \eta)^2}, \quad (9)$$

where $\eta = \pi n \sigma^2 / 4$ is the packing fraction, $n = N/V$ is the number density with N being the number of particles in the system and V is the two-dimensional volume (area). The Enskog factor obviously accounts only for static spatial correlations. Any dynamic correlations between the velocities of particles are still ignored.

For the case of dissipative gases there is an additional source of correlations. These arise since the restitution coefficient affects only the normal component of the relative velocity of a colliding pair, leaving the tangential relative velocity unchanged. If two inelastic particles collide with a (pre-collision) angle α'' the angle α after the collision will be smaller, $\alpha < \alpha''$ (see Fig. 1).

As a consequence subsequent collisions lead to more and more aligned traces of neighboring particles. It has been shown^{6,13} that these correlations give rise to convection cells in an otherwise homogeneous granular gas, i.e., to macroscopic correlations in the velocity field even in absence of spatial inhomogeneities.

Since the particles are aligned in macroscopic vortices, such velocity correlations lead to a reduced collision frequency as compared with the collision frequency of a gas of elastic particles of the same temperature. The reduction of the collision frequency leads in its turn to a reduction of the cooling rate, i.e., to a retarded decay of temperature even in the stage of homogeneous cooling. Therefore, to estimate

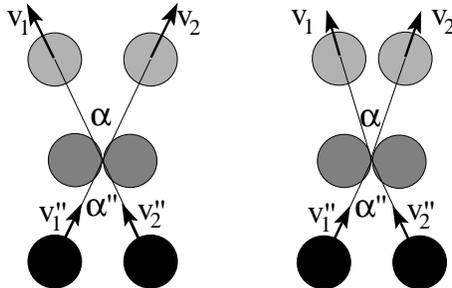


Fig. 1. Elastic (left) and inelastic (right) spheres before (black), during (dark grey) and after a collision (light grey). For elastic spheres the incoming angle equals the outgoing angle ($\alpha = \alpha''$) whereas for inelastic collisions $\alpha < \alpha''$.

the importance of velocity correlations we analyze the cooling rate of a dissipative gas.

4. Temperature decay of dissipative gases

Assume a homogeneous dissipative gas of particles with velocities which obey a distribution function $f(\vec{v})$. For $\epsilon = \text{const.}$ the Boltzmann equation decouples into a set of simpler equations¹⁴, one for the time dependent temperature

$$T(t) \equiv \frac{m}{3n} \int d\vec{v} v^2 f(\vec{v}, t) \quad (10)$$

and another one for the time independent (normalized) distribution function. With the assumption that the velocities obey a Maxwell distribution it has been shown by scaling arguments that temperature decays due to dissipative collisions as

$$T(t) = \frac{T_0}{(1 + t/\tau_0)^2} \quad (11)$$

with T_0 being the initial temperature and τ_0 being a scaling factor which will be discussed in detail below. Equation (11) is known as Haff's law¹⁵. Later deviations from the Maxwell distribution have been found^{16,17}, however, the temperature decay law is still correct.

Let us compare the analytical results for the temperature evolution with the temperature decay as obtained by an event driven Molecular Dynamics simulation. Haff's law was derived with the molecular chaos assumption, therefore, in order to obtain results which are comparable with theory we apply a special numerical procedure which destroys the collision-induced velocity correlation of colliding particles. This procedure is illustrated in Fig. 2. We shuffle the velocities of the colliding particles with the velocities of randomly selected particles. It is easy to understand that the shuffling procedure is unique since the condition for the after-collision normal relative velocity to be positive must be fulfilled.

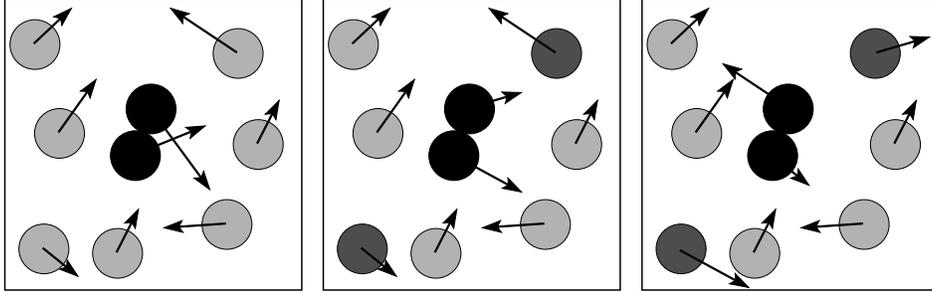


Fig. 2. The velocity shuffling procedure. Left: the black particles at the instant of the collision with pre-collision velocities. Middle: the colliding particles are assigned new velocities due to the collision rule Eq. (5), two randomly selected particles are drawn dark grey. Right: the velocities of the colliding particles are shuffled with the velocities of the randomly selected particles. The shuffling procedure is unique due to the condition for the after-collision normal relative velocity to be positive.

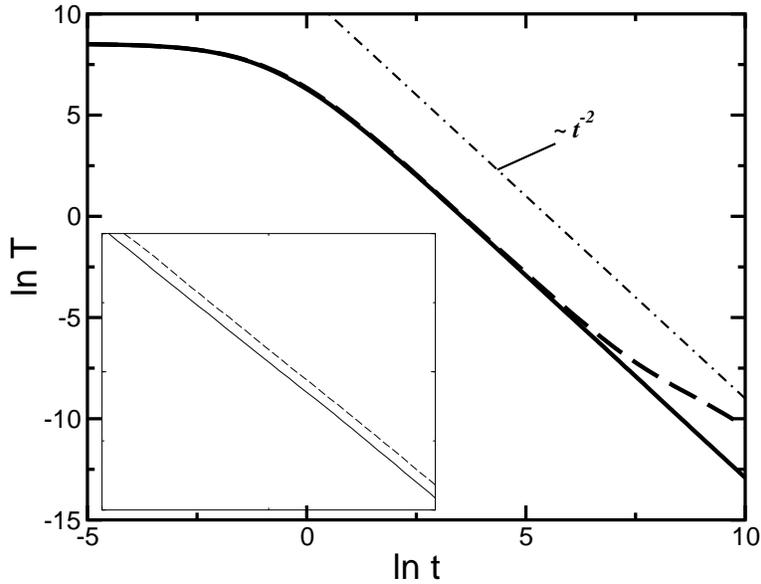


Fig. 3. Evolution of the temperature. Dashed line: event driven MD simulation ($\epsilon = 0.93$, $n = 0.05$), full line: Haff's law, Eq. (11) together with the randomized event driven simulation (lines fall together). As soon as clusters appear ($\ln t \approx 6$) there is a large deviation of the numerical results from theory. The inset shows a magnification of the range $2.5 \leq \ln t \leq 3$. In the entire range of time we observe a small but systematic deviation between the curves.

The solid line in Fig. 3 shows the evolution of temperature for a system of $N = 5,000$ inelastically colliding spheres in two dimensions with periodic boundary conditions where the described shuffling procedure has been applied (randomized simulation). The reduced density was $n = N\sigma^2/V = 0.05$ (with $\sigma = 1$), where V is the area of the quadratic box and the coefficient of restitution is $\epsilon = 0.93$. In the entire range of t the curve is identical up to the line width with Haff's law (11) where the relaxation time τ_0 is a fit parameter. Repeating the same simulation without the shuffling procedure we obtain a temperature decay as drawn by the dashed line in Fig. 3. The curves are close to each other until $\ln t \approx 6$. At this time instant for the unshuffled system we observe formation of spatial inhomogeneities (clusters) which leads to a reduced collision frequency. The cluster formation in dissipative gases due to a pressure instability is a well understood phenomenon which was described first in¹⁸.

Obviously, our randomization procedure suppresses the formation of density inhomogeneities which can be understood from mode-coupling analysis.¹⁸ We know that the space inhomogeneities arise in two steps: first vortex patterns appear due to interparticle velocity correlations induced by the collisions, then clusters develop owing to interaction between the density modes and the flow-velocity modes. Thus, the collision-induced correlations play a clue role in the formation of spatial inhomogeneities. Therefore, since clustering or vertex formation is not possible in the randomized simulation due to suppression of the collision-induced velocity correlations we find almost perfect agreement with Haff's law.

Figure 3 shows that in the region $\ln t \lesssim 6$ Haff's law describes the temperature decay of the dissipative gas with and without randomization up to a good degree of accuracy. Having a closer look to Fig. 3 (inset) one recognizes, however, a small but systematic deviation between these two temperature curves. We have checked spatial homogeneity of the particle distribution for the both gases and did not observe any difference in the time span where Haff's law is applicable. Thus we conclude that this difference may be attributed only to velocity correlations of the particles of the gas.

It is possible to fit the value of the relaxation time τ_0 in Haff's law to reproduce the simulation data (dashed line) up to the same accuracy as done before for the randomized gas. The numerical value of τ_0 which depends on the coefficient of restitution and on the density of the gas may, therefore, serve to characterize the velocity correlations of the particles. We will return to this issue in Sec. 6.

5. Beyond the concept of molecular chaos

To develop a theory beyond the concept of molecular chaos we introduce the reduced velocities

$$\vec{c} \equiv \frac{\vec{v}}{v_0} \quad \text{with} \quad v_0 \equiv \sqrt{\frac{2T}{m}}, \quad (12)$$

where v_0 is the thermal velocity and the reduced velocity distribution function $\tilde{f}(\vec{c})$

which is defined by

$$f(\vec{v}, t) = \frac{n}{v_0^3(t)} \tilde{f}(\vec{c}, t). \quad (13)$$

In the equivalent way we define the reduced two-particle distribution function $\tilde{f}_2(\vec{c}_1, \vec{c}_2, \vec{r}_{12}, t)$ with $\vec{r}_{12} \equiv \vec{r}_1 - \vec{r}_2$.

Correlations of the particle velocities are taken into account by assuming the following form of the two-particle velocity distribution function:

$$\tilde{f}_2(\vec{c}_1, \vec{c}_2, \vec{r}_{12}, t) = \tilde{f}(c_1) \tilde{f}(c_2) \left[g_2(r_{12}) + \chi(r_{12}) \hat{D}(\hat{r}_{12}) : \vec{c}_1 \vec{c}_2 \right] \quad (14)$$

The first term in the brackets in the right-hand side of Eq. (14) accounts only for static correlations, while the second term accounts for dynamic correlation of a pair of particles with the reduced velocities \vec{c}_1 and \vec{c}_2 . The traceless dyad of the unit vector $\hat{r}_{12} = \vec{r}_{12}/r_{12}$ is defined as

$$\hat{D}(\hat{r}_{12}) \equiv 3 \hat{r}_{12} \hat{r}_{12} - \hat{U} \quad (15)$$

with \hat{U} being the unit tensor and we use the conventional notation of vector algebra, i.e., $\vec{a}\vec{a}$ denotes the dyad and

$$\hat{D}(\hat{r}_{12}) : \vec{c}_1 \vec{c}_2 \equiv 3 (\vec{c}_1 \cdot \hat{r}_{12}) (\vec{c}_2 \cdot \hat{r}_{12}) - (\vec{c}_1 \cdot \vec{c}_2). \quad (16)$$

This is one of the simplest forms which may be built up from three vectors and which features the requested symmetry with respect to vectors \vec{c}_1 and \vec{c}_2 . The function $\chi(r_{12})$ expresses the dependence of the velocity correlations on the particle distance which, obviously, vanishes for infinite distance, i.e., $\chi(r_{12}) \rightarrow 0$ for $r_{12} \rightarrow \infty$.

At the instant of a collision, i.e., when $r_{12} = \sigma$ (or $\hat{r}_{12} = \vec{e}$) Eq. (14) reads

$$\tilde{f}_2(\vec{c}_1, \vec{c}_2, \sigma \vec{e}) = \tilde{f}(c_1) \tilde{f}(c_2) g_2(\sigma) \{1 + b [3 (\vec{c}_1 \cdot \vec{e}) (\vec{c}_2 \cdot \vec{e}) - (\vec{c}_1 \cdot \vec{c}_2)]\}, \quad (17)$$

where $b \equiv \chi(\sigma)/g_2(\sigma)$. For the one-particle distribution function we use its representation in terms of the Sonine polynomial expansion with the first non-vanishing Sonine coefficient a_2 :

$$\tilde{f}(\vec{c}_1) = \phi(c) [1 + a_2 S_2(c^2)], \quad (18)$$

where for the two-dimensional case the second Sonine polynomial reads,

$$S_2(x) = \frac{1}{2} x^2 - 2x + 1. \quad (19)$$

With the distribution function given by Eqs. (17,18,19) all necessary quantities which describe the kinetic properties of the gas can be obtained, e.g., the second moment of the collision integral

$$\mu_2 = -\frac{1}{2} \int d\vec{c}_1 \int d\vec{c}_2 \int d\vec{e} \Theta(-\vec{c}_{12} \cdot \vec{e}) |\vec{c}_{12} \cdot \vec{e}| \tilde{f}_2(\vec{c}_1, \vec{c}_2, \sigma \vec{e}) \Delta(c_1^2 + c_2^2), \quad (20)$$

where $\Delta(c_1^2 + c_2^2)$ denotes the change of the quantity $c_1^2 + c_2^2$ at the dissipative collision.

The importance of the second moment μ_2 follows from the fact that it characterizes the dissipation rate due to inelastic collisions. Indeed, the factor $\Theta(-\vec{c}_{12} \cdot \vec{e})$ in the integrand guarantees that particles with velocities \vec{c}_1 and \vec{c}_2 do collide. The factor $|\vec{c}_{12} \cdot \vec{e}|$ characterizes the length of the collision cylinder, which multiplied by the factor $\tilde{f}_2(\vec{c}_1, \vec{c}_2, \sigma\vec{e})$ characterizes the number of collisions between particles with velocities \vec{c}_1 , \vec{c}_2 and the intercenter vector $\sigma\vec{e}$, occurring per unit time. The last factor in the integrand $\Delta(c_1^2 + c_2^2)$ characterizes the energy loss in such collisions. Integration over all possible velocities and intercenter vectors yields the quantity which characterizes the energy loss per unit time, i.e., the decay rate of the gas temperature.

Straightforward calculations yield

$$\mu_2 = \frac{1}{2}\sqrt{2\pi}(1 - \epsilon^2) \left[1 + \frac{3}{16}a_2 - \frac{63}{40}b \right]. \quad (21)$$

This expression shows that μ_2 depends on both coefficients a_2 and b in a similar way.

6. Influence of the velocity correlations on the temperature evolution

Originally, Haff's law (11) was derived by scaling arguments with the assumption of the Maxwell distribution for the particle velocities.¹⁵ From this derivation it is not possible to obtain the relaxation time τ_0 in terms of the microscopic particle properties. Starting with the Boltzmann-Enskog equation, however, the temperature evolution¹⁷, i.e. Haff's law, and expression for the relaxation time τ_0 in terms of the second moment of the collision integral can be found:

$$\tau_0^{-1} = \frac{1}{3}g_2(\sigma)\sigma^2 n \sqrt{\frac{2T_0}{m}} \mu_2. \quad (22)$$

With expression (21) for μ_2 we obtain

$$\tau_0 = \frac{1}{1 - \epsilon^2} A(\epsilon), \quad (23)$$

with

$$A^{-1}(\epsilon) = \omega_0 \left[1 + \frac{3}{16}a_2(\epsilon) - \frac{63}{40}b(\epsilon) \right], \quad \omega_0 = \frac{1}{3}g_2(\sigma)\sigma^2 n \sqrt{\pi \frac{T_0}{m}}. \quad (24)$$

The second Sonine coefficient for a two-dimensional gas of inelastic discs as a function of the coefficient of restitution has been obtained with the molecular chaos assumption (i.e. ignoring particle correlations)¹⁶:

$$a_2 = \frac{16(1 - \epsilon)(1 - 2\epsilon^2)}{57 - 25\epsilon + 30\epsilon^2(1 - \epsilon)}. \quad (25)$$

Using Eqs. (23,24,25) the (macroscopic) relaxation time τ_0 is expressed in terms of microscopic quantities which characterize the particle collisions.

Analyzing Eqs. (24,25) it turns out that the dependence of $A(\epsilon)$ on ϵ is very weak: In the interval $\epsilon \in (0.8, 1.0)$ it varies within less than 0.6%. From Eq. (24) follows that the velocity correlations, expressed by b preserve the functional form of the temperature decay, but change the value of the relaxation time. Moreover, it also follows that the more pronounced the velocity correlations (i.e. the larger the coefficient b) the more the relaxation time τ_0 differs from the relaxation time of a gas where the velocity correlations are suppressed. Taking into account that a_2 is small and that b is also not large, we can write for the factor $A(\epsilon)$:

$$A = \omega_0^{-1} \left[1 - \frac{3}{16} a_2(\epsilon) + \frac{63}{40} b(\epsilon) \right] = A_{\text{rand}} + \frac{63}{40} \omega_0^{-1} b(\epsilon) \quad (26)$$

where A_{rand} is the factor A that corresponds to a gas without velocity correlations, i.e., a randomized gas.

This theoretical prediction can be checked by numerical simulations: again we perform event driven Molecular Dynamics simulations, measure the temperature as a function of time and determine the value of τ_0 as a fit parameter due to Eq. (11). According to Eq. (23) the function $A(\epsilon)$ can be obtained by drawing $\tau_0 (1 - \epsilon^2)$ over ϵ . The results are shown Fig. 4 for two values of the density n .

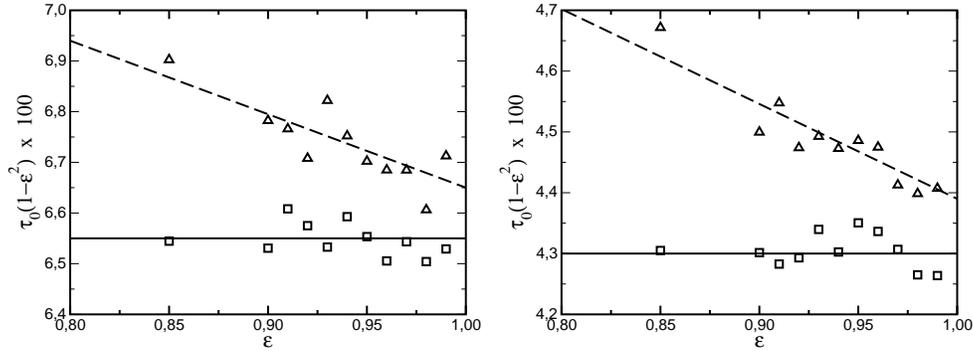


Fig. 4. The function $A(\epsilon) \equiv \tau_0 (1 - \epsilon^2)$ over the coefficient of restitution ϵ for a simulation of 5,000 inelastic hard disks of density $n = 0.05$ (left) and $n = 0.1$ (right). The lines have been fit to the numerical data. For the randomized simulation (squares) the curve does not significantly depend on ϵ whereas in agreement with the theoretical prediction Eq. (26) the original gas shows noticeable dependence on ϵ due to velocity correlations.

The squares corresponds to the randomized gas and do not exhibit any noticeable dependence on the coefficient of restitution ϵ . As it has been already mentioned this is a consequence of the very weak dependence of A on ϵ via the second Sonine coefficient a_2 . The triangles show the same data for simulations where the randomizing procedure has not been applied. The lines show linear least square fits to the

numerical data. Figure 4 shows that the dependence of A on ϵ via the correlation coefficient b can be approximated by a linear function of ϵ .

7. Conclusions

We study microscopic particle velocity correlations in dissipative gases. Taking into account velocity correlations preserves the functional form of the temperature decay, which still obeys Haff's law but changes the temperature relaxation time. Going beyond the assumption of molecular chaos we suggest the functional form of the two-particle distribution function which accounts for velocity correlations and calculate the temperature relaxation time for this distribution function. We performed Molecular Dynamics simulations of dissipative gases and suggest a randomizing procedure which suppresses the velocity correlations but leaves all other properties of the gas dynamics unchanged. From the comparison of the results of the simulation of the randomized gas with traditional simulations we obtain the dependence of the correlation coefficient on the coefficient of restitution.

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