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Violation of the Einstein relation in granular fluids: the role of correlations

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Abstract. We study the linear response in different models of driven granular gases. In some situations, even if the velocity statistics can be strongly non-Gaussian, we do not observe appreciable violations of the Einstein formula for diffusion versus mobility. The situation changes when strong correlations between velocities and density are present: in this case, although a form of fluctuation-dissipation relation holds, the differential velocity response of a particle and its velocity self-correlation are no longer proportional. This happens at high densities and strong inelasticities, but still in the fluid-like (and ergodic) regime.

Keywords: transport processes/heat transfer (theory), granular matter, Brownian motion, fluctuations (theory)

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Contents

1. Introduction

The transport properties of flowing dilute granular materials constitute an open problem in non-equilibrium statistical mechanics [1, 2]. The existing kinetic theories aiming to deduce transport properties from the microscopic dynamics for the 'usual' gases have a hard life, here, because of the presence of inelastic interactions among grains, that prevent the assumption of an equilibrium measure in the unperturbed state. Several approaches to this problem have been proposed in previous studies.

In a large series of works, there has been considered a set-up where energy is injected only through the boundaries of the system. In this treatment, the bulk is considered as a 'freely evolving' inelastic gas, which would cool down if not driven by energy currents transported by the gas itself. It is possible to write down balance equations for local fields such as density, velocity and kinetic temperature and, through a very delicate assumption of separation between microscopic and mesoscopic scales [3], the so-called granular hydrodynamics can be obtained [4]. Following these lines, fluctuation response relations have been obtained with respect to the homogeneous cooling state, i.e. relaxation laws for small perturbations of a state whose fate is thermal death [5, 6].

A different approach consists in considering an alternative experimental set-up where the starting state is much more similar to a thermal state, such as the thermodynamic equilibrium of a gas. Such a state can be prepared by coupling the energy source to all grains of the system, for example in granular materials fluidized by some air flow [7], or otherwise in granular beds put on a vibrating plate [8, 9]. In such cases, the unperturbed fluid state is stationary and one can study how the system relaxes to it when a small perturbation is applied. Models for these granular stationary states have been proposed [10]–[12], showing the main differences with respect to the thermal state of a molecular gas: lack of equipartition, departure from Gaussian statistics of velocities, tendency to enhance spatial grain–grain correlations, clustering. It must be remarked that the obtained stationary state is intrinsically out of equilibrium: a net current of energy flows from the external source, through inelastic collisions, into heat [13, 14]. A Boltzmann equation for such a class of granular fluids has been proposed [15], as well as a kinetic theory for transport coefficients [16, 17]: these theories have their validity in a suitable range of the parameters. Recently numerical studies have been performed showing that, in homogeneous situations, the fluctuation response relation (FR) is valid in its near-equilibrium formulation, replacing the bath temperature with the internal granular temperature [18, 19]. This has interesting consequences in the case of mixtures, where different components have different temperatures [20]: for instance, a linear response experiment on a massive tracer, performed to obtain a temperature measurement (a granular thermometer), yields the temperature of the tracer and not that of the surrounding gas. The verification of FR has been explained by means of a hydrodynamic approach by Garzo [21], who connected it to the very small departures from the Maxwell–Boltzmann statistics. Other studies on the FR in models of granular systems have shown some deviations from the Einstein formula [22]. In the following, we discuss when this particular kind of FR ceases to be valid in a driven granular system: it will appear that the most relevant ingredient is not the deviation from the Maxwell–Boltzmann statistics, but the degree of correlations among different degrees of freedom (d.o.f.), which increases as the total excluded volume decreases.

The aim of this paper is to put this problem in the more general context of linear response theory for statistically stationary states, whose formulation has been given in [23, 24] and which can be described in very general terms. Consider a dynamical system $\mathbf{X}(0) \to \mathbf{X}(t) = U^t\mathbf{X}(0)$ whose time evolution can also be not completely deterministic (e.g. stochastic differential equations), with states $\mathbf{X}$ belonging to an N-dimensional vector space. We assume (a) the existence of an invariant probability distribution $\rho(\mathbf{X})$, for which an 'absolute continuity' condition is required, and (b) the mixing character of the system (from which its ergodicity follows). These assumptions imply also that the system is time translation invariant (TTI). Now we introduce the two main ingredients of the theory: the response of the system to a small perturbation, and the time correlation of the unperturbed system that describes the relaxation of its spontaneous fluctuations. In the following we will indicate with $\langle\cdot\rangle$ an average in the unperturbed system, i.e. weighting states with the invariant measure, and with $\overline{(\cdot)}$ the time dependent average in the dynamical ensemble generated by the external perturbation.

In the unperturbed system, the relaxation of spontaneous fluctuations is described by the time dependent cross correlations of two generic observables $A(\mathbf{X})$ and $B(\mathbf{X})$

$$C_{AB}(t) = \langle A(\mathbf{X}(t))B(\mathbf{X}(0))\rangle. \tag{1}$$

The average effect at time t on a variable X_i of a small external perturbation $f(s)$, for instance on the variable X_j, applied at time s can be written, in the linear response regime, as

$$\overline{\delta X_i(t)} = \int_0^t R_{i,j}(t-s)f(s)\,\mathrm{d}s, \tag{2}$$

which defines the response function $R_{i,j}(t)$. The basic idea of the FR is to link the response functions $\{R\}$ to suitable correlations $\{C\}$ of the unperturbed system.

For example, let us consider a colloidal particle in a fluid with friction constant γ and temperature T: one has a system with two d.o.f., position and velocity of the particle,

(X, V), whose evolution is given by the Langevin equation:

$$\frac{\mathrm{d}X}{\mathrm{d}t} = V \tag{3}$$

$$\frac{\mathrm{d}V}{\mathrm{d}t} = -\gamma V + \sqrt{\frac{2\gamma T}{M}}\eta, \tag{4}$$

where η is a white noise, i.e. a Gaussian stochastic process with $\langle \eta(t) \rangle = 0$ and $\langle \eta(t)\eta(t') \rangle = \delta(t - t')$ (for the sake of simplicity we will assume the Boltzmann constant $k_{\mathrm{B}} = 1$). The time self-correlation of the particle velocity, $C_{VV}(t) = \langle V(t)V(0) \rangle = \langle V^2 \rangle \mathrm{e}^{-\gamma t}$, when integrated from 0 to ∞, determines the self-diffusion coefficient: $D = \int_0^\infty \langle V(t)V(0) \rangle \, \mathrm{d}t$. It describes the asymptotic growth of the mean square displacement of the particle: $\langle (X(t) - X(0))^2/t \rangle \to 2D$. On the other hand, when the momentum of the particle is perturbed with a force $f(t) = F\Theta(t)$, where $\Theta(t)$ is the Heaviside step function, the response of the velocity itself, at very large times, reads

$$\overline{\delta V(\infty)} = \frac{F}{M} \int_0^\infty R(t) \, \mathrm{d}t = \mu F, \tag{5}$$

which defines the mobility μ. An easy computation gives

$$\mu = \beta D, \tag{6}$$

where $\beta = 1/T$. This relation, well known as the Einstein formula, obtained in his celebrated 1905 paper on Brownian motion [25, 26], is a primordial example of the fluctuation response relation: it relates, in fact, the response to a perturbation to the relaxation of spontaneous fluctuations.

After the publication of the Einstein relation, a large amount of work [27]–[31] was devoted to generalizing it to the class of (classical as well as quantal) Hamiltonian systems coupled to a thermostat at temperature T. This means considering dynamical systems with variables $(\mathbf{q}, \mathbf{p})$ whose time evolution is generated by a Hamiltonian $\mathcal{H}_0$. The external perturbation appears as a perturbation of the Hamiltonian $\Delta\mathcal{H}(t) = f(t)B_f(\mathbf{q}, \mathbf{p})$. In this case, the fluctuation response relation can be written in, among others, the following form:

$$\frac{\overline{\delta A(t)}}{\delta f(s)} = R_{A,f}(t) = \beta \langle A(t)\dot{B}_f(s) \rangle. \tag{7}$$

The response of the observable A at time t with respect to a 'force' f applied to the system at time s is related to the correlation, measured in the unperturbed system, between the observable itself at time t and the time derivative of the observable B_f at time s. The latter is the one conjugated to f through the Hamiltonian.

The fact that the FR theory was developed in the context of equilibrium statistical mechanics of Hamiltonian systems generated some confusion and misleading ideas on its validity. As a matter of fact it is possible to show that a generalized FR relation holds under the rather general hypotheses discussed above, i.e. basically the mixing property and the existence of an absolute continuous invariant measure $\rho(\mathbf{X})$. The main result (for details see [24]) is the following fluctuation response relation, valid when considering the

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perturbation at time 0 of a coordinate X_j:

$$R_{i,j}(t) = \frac{\overline{\delta X_i(t)}}{\delta X_j(0)} = -\left\langle X_i(t) \frac{\partial \ln \rho(\mathbf{X})}{\partial X_j}\bigg|_{t=0}\right\rangle. \tag{8}$$

From this relation all previous cases can be obtained. The Brownian motion of the colloidal particle, for example, has an invariant measure where position and velocity are independent: the part concerning V is of course a Gaussian with $\langle V \rangle = 0$ and $\langle V^2 \rangle = T/M$. From formula (8), therefore, it follows that

$$R_{V,V} = M\beta \langle V(t)V(0)\rangle \tag{9}$$

and this immediately returns the Einstein relation (6). In the rest of the paper, with a slight abuse of terminology, we will use the form 'Einstein relation' to denote the time dependent form (9).

In the case of thermostatted Hamiltonian systems, on the other side, one has that $\rho(\mathbf{q},\mathbf{p}) \propto \exp(-\beta\mathcal{H}(\mathbf{q},\mathbf{p}))$. In such a case equation (8) gives for example

$$R_{p_i,p_i}(t) = \beta\left\langle p_i(t)\frac{\partial \mathcal{H}}{\partial p_i}\bigg|_{t=0}\right\rangle = \beta\left\langle p_i(t)\frac{\mathrm{d}}{\mathrm{d}t}q_i(0)\right\rangle = -\beta\frac{\mathrm{d}}{\mathrm{d}t}\langle p_i(t)q_i(0)\rangle. \tag{10}$$

In non-Hamiltonian (and in general non-Gaussian) systems, the shape of $\rho(\mathbf{x})$ is not known; therefore (8) does not give straightforward information. However from it one can see that a FR relation still exists, stating the equivalence of the response to a suitable correlation function computed in the non-perturbed systems. This means that from an ansatz on the invariant measure ρ one can directly deduce the response matrix.

Following these lines, we analyse the response to small perturbations of a thermostatted granular gas, trying to connect the response properties of the stationary state with its many 'anomalies' with respect to an equilibrium state. In particular we show (section 2) that, in homogeneous situations, even when the invariant measure is far from the Gaussian, the Einstein relation holds with good accuracy. In contrast (section 3), when the granular effects (excluded volume and inelasticity) are strong enough to develop correlations between local density and velocities, the invariant measure of the system becomes highly non-trivial, and the Einstein relation is no longer observed. We stress that the regimes considered here are always ergodic: this is a relevant difference with respect to previous studies on the violations of the fluctuation response relation, which considered glassy systems in the non-ergodic (ageing) phase [32].

2. Homogeneous granular fluids

2.1. The models

We start by discussing the linear response of a dilute granular gas with N grains of mass $m = 1$. Three different models, all in dimension $d = 2$, are considered here:

(i) the homogeneously driven gas of inelastic hard discs in the dilute limit, evolving through stochastic molecular dynamics rules;

(ii) the homogeneously driven gas of inelastic hard discs in the molecular chaos approximation, i.e. where its dynamics is determined by the homogeneous direct simulation Monte Carlo (DSMC) algorithm;

(iii) the homogeneous inelastic Maxwell model driven by a 'Gaussian thermostat'.

In all the above models one has

$$\rho(\{\mathbf{v}_i, x_i\}) = n^N \prod_{i=1}^{N} \prod_{\alpha=1}^{d} p_v(v_i^{(\alpha)}) \tag{11}$$

with n the spatial density $n = N/V$ and $p_v(v)$ the one-particle velocity component probability density function, $v_i^{(\alpha)}$ the αth component of the velocity of the ith particle and d the system dimensionality. In particular, in models 2 and 3 this is true by assumption, while for model 1 it is well verified in simulations, as a consequence of being dilute. In view of the fact that all discussed models are isotropic, in the following we will denote with v an arbitrary component of the velocity vector: the results do not change if v is the x or y component.

The three models are known to display non-Gaussian $p_v(v)$. From the above discussion, it is expected that an instantaneous perturbation $\delta v(0)$ at time $t = 0$ of a particle of the gas will result in an average response of the form

$$R(t) = \frac{\overline{\delta v(t)}}{\delta v(0)} = -\left\langle v(t) \frac{\partial \ln p_v(v)}{\partial v}\bigg|_0 \right\rangle \neq C_1(t), \tag{12}$$

where we have defined $C_1(t) = \langle v(t)v(0)\rangle / \langle v^2 \rangle$.

Some previous studies already showed that for the inelastic hard discs model, in the dilute limit, it is very difficult to observe the discrepancy between $R(t)$ and $C_1(t)$, i.e. to see any 'violation' of the Einstein relation for mobility and diffusion. In such studies, however, the deviation from a Gaussian $p_v(v)$ was always small, i.e. consistent with a Sonine polynomial fit with a parameter $a_2 = \langle v^4 \rangle / 3\langle v^2 \rangle^2 - 1 \ll 1$ [15]. Actually, in a driven dilute system it is very rare to observe large departures from the Gaussian behaviour. On the other hand, studying models such as the thermostatted Maxwell model, or tuning the parameters of the DSMC algorithm for inelastic hard discs beyond the dilute limit, one can induce rather large deviations from the Gaussian, while condition (11) still holds. Even if this may be far from being realistic, it is useful for assessing the relevance of the non-Gaussian velocity pdf for the linear response of the gas.

For the three models, the unperturbed dynamics is determined by a non-interacting streaming (where each particle is coupled to the thermostat only) plus inelastic collisions. For model (i), which is the closest to experimental results for driven granular gases, the streaming part is described by the equations of motion of N hard discs of diameter $\sigma = 1$ moving in a square of area $V = L \times L$ with periodic boundary conditions and coupled to a thermal bath with viscosity γ and temperature T_{b}:

$$\frac{\mathrm{d}\mathbf{x}_i(t)}{\mathrm{d}t} = \mathbf{v}_i(t) \tag{13}$$

$$\frac{\mathrm{d}\mathbf{v}_i(t)}{\mathrm{d}t} = -\gamma \mathbf{v}_i(t) + \sqrt{2\gamma T_{\mathrm{b}}}\,\boldsymbol{\eta}_i(t), \tag{14}$$

with $\boldsymbol{\eta}$ a Gaussian white noise, i.e. $\langle \eta_i^{(\alpha)}(t) \rangle = 0$ and $\langle \eta_i^{(\alpha)}(t) \eta_j^{(\alpha')}(t') \rangle = \delta_{\alpha,\alpha'} \delta_{ij} \delta(t - t')$, where α and α' indicate the Cartesian components. When two grains i and j touch, an instantaneous inelastic collision takes place, with a change of velocities given by

$$\mathbf{v}_i' = \mathbf{v}_i - \frac{1+r}{2}[(\mathbf{v}_i - \mathbf{v}_j) \cdot \boldsymbol{\sigma}]\boldsymbol{\sigma} = \mathbf{v}_i + \Delta \mathbf{v}_{i,\mathrm{col}}(\mathbf{v}_i, \mathbf{v}_j, \boldsymbol{\sigma}) \tag{15}$$

where $r \in [0, 1]$ is the restitution coefficient (the elastic case corresponds to $r = 1$) and $\boldsymbol{\sigma}$ is the unit vector joining the centres of the two colliding particles. In the dilute limit, numerical simulations show that colliding particles are not correlated, i.e. molecular chaos holds. The system is known to display very different regimes when τ_b/τ_c changes, where $\tau_b = 1/\gamma$ and $\tau_c = 1/\omega_c$ is the average mean free time between collisions of a single particle. If $\tau_b/\tau_c \gg 1$, the effect of collisions is very small and, even if inelastic, the gas behaves as at equilibrium at temperature T_b. In the opposite case, $\tau_b/\tau_c \ll 1$, collisions are dominant and the gas reaches a fluctuating stationary state with 'granular temperature' $T_g \equiv \langle |v|^2 \rangle/2 < T_b$ (the smaller r, the smaller T_g). In this non-equilibrium regime, the velocity pdf is non-Gaussian with slow tails at very large $|v|$. Note that, here, τ_c is not an external parameter, but is self-determined by the system: increasing $n\sigma$, i.e. reducing the mean free path, results in a smaller granular temperature and the same happens when increasing τ_b; therefore in both cases a direct increase of τ_b/τ_c is not obvious. Direct experience teaches us that, when diluteness (volume fraction $\phi = n\pi\sigma^2/4 \ll 1$) is also required, then it is very difficult to obtain $\tau_c \ll \tau_b$. For instance, when $\phi \sim 0.1$, we usually observe $\tau_c \sim 0.1\tau_b$ or larger.

Model (ii) consists of the same physical system with the enforcement of both spatial homogeneity and molecular chaos. This is achieved by disregarding the spatial coordinates of the particles, and selecting with a stochastic rule the pairs of particles involved in each collision. Time is discretized in small steps of length Δt (smaller than τ_b and τ_c). At each step the discretized version of (14) is used to evolve the velocities of all the particles. Then, a number of collisions $N\omega_c\Delta t/2$ are performed, where $\omega_c = 2\sigma n\sqrt{\pi T_g}$ is the theoretical one-particle collision frequency for a dilute gas. Pairs i, j to collide are chosen with a probability proportional to the quantity $-(\mathbf{v}_i - \mathbf{v}_j)\cdot\boldsymbol{\sigma}\Theta(-(\mathbf{v}_i - \mathbf{v}_j)\cdot\boldsymbol{\sigma})$, with $\boldsymbol{\sigma} = (\cos\theta, \sin\theta)$ and θ chosen randomly with uniform probability in $[0, 2\pi)$. This process mimics the relative velocity dependent collision frequency in dilute gas of particles with hard core interactions. Since, typically, a particle i, after having collided with a particle j, will have a second collision with the same particle j after a number of collisions of order N, any memory of the first collision will be lost and the new collision can be considered uncorrelated with the previous one. The phenomenology observed in this model is analogous to that of model (i) in its dilute limit. On the other side, here one can arbitrarily tune the ratio τ_b/τ_c, increasing $n\sigma$ and ignoring the inconsistency between the high density and the enforced molecular chaos.

Finally, model (iii), the inelastic Maxwell model, is a further simplification of model (ii): couples of particles collide with a constant probability, i.e. independently of the relative velocity. The collision frequency is assumed to be $\omega_c = 1/N$. Moreover, the streaming part of the dynamics is performed with $T_b = 0$ and $\gamma = -\lambda$ with $\lambda = (1-r^2)/4$, i.e. negative friction and no random forces. This is the so-called Gaussian thermostat, which guarantees a constant kinetic energy, since in the non-driven case ('free cooling') the total energy of the system would decay as $\sim\exp(-\lambda t)$. It has been shown that such a 'thermostat' is equivalent to considering the free cooling system and continuously rescaling all particle velocity components by a factor $\sqrt{T_g}$. The analysis of this idealized granular model is instructive for the following reasons. First, it has been shown [33] that such a model has a stationary probability density function for the velocity bv with power law tails. In particular, it displays $p_v(v)$ with high energy tails of the form v^{-b} with $b = 4$ in $d = 1$ and $b > 4$ in $d = 2$ (a good estimate for not too high inelasticity is $b \simeq 4/(1-r)$).

Second, the simplification of the dynamics allows a direct analytical computation of time correlations and responses.

2.2. The numerical experiment

The protocol used in our numerical experiment, for the three models, is the following:

- First, the gas is prepared in a 'thermal' state, with random velocity components extracted from a Gaussian with zero average and variance $T_g(0)$. Positions of the particles, relevant only in model (i), are chosen uniformly random in the box, avoiding overlapping configurations.
- Second, the system is let evolve until a statistically stationary state is reached, which is set as time zero: we verify that the total kinetic energy fluctuate around an average steady value and that this value does not depend upon initial conditions. In the case of model (iii) the energy is stationary by definition; therefore we ensure that a stationary velocity pdf is observed.
- Third, a copy of the system is obtained, identical to the original but for one particle, whose x (for instance) velocity component is perturbed of an amount $\delta v(0)$.
- Finally both systems are let evolve with the unperturbed dynamics. In models (i) and (ii), which involve random thermostats, the same noise is used. After a time $t_{\max}$ large enough for having lost memory of the configuration at time zero, a new copy is taken, perturbing a new random particle and repeating the response measurement.

This procedure is performed many times, in order to reduce the statistical errors for both the response and all required self-correlations in the unperturbed copy. In the following, averages indicated as $\langle \cdot \rangle$ and $\overline{(\cdot)}$ will have the meaning of averages over many realizations of this procedure.

In figure 1 we show the results of these experiments for the three different models. In the right frame the velocity pdfs are shown for different choices of the parameters in those models. For molecular dynamics simulations of inelastic hard discs, even if quite inelastic, but still dilute ($\phi = n\pi\sigma^2/4 < 0.1$), the velocity pdf is not very far from a Gaussian, as in a DSMC with similar choices of the parameters ($\tau_c \sim 0.1\tau_b$). Increasing n in the DSMC leads to stationary regimes very far from thermal equilibrium, with $T_g \ll T_b$ and larger tails of the velocity pdf $p_v(v)$, with $a_2 \sim 0.1$. In view of relation (12), we have tried a three-parameter fitting of the kind

$$p_v(v) = c_0 \exp(-c_1 v^2 + c_2 |v^3| + c_3 v^4) \tag{16}$$

where c_0 is not independent because of normalization. In most of the observed cases $|c_3 v_{\max}^4| \ll |c_2 v_{\max}^3|$, with $v_{\max}$ the largest value of v in the histogram. Therefore for our aims, in practice, we can drop the (negative) quartic term, retaining only c_1 and c_2. The obtained fit appears to be very good; see figure 1. Using this formula in equation (12), we get

$$R(t) = -2c_1 \langle v(t) v(0) \rangle + 3c_2 \langle v(t) |v(0)| v(0) \rangle \tag{17}$$

$$= -2c_1 \langle v^2 \rangle C_1(t) + 3c_2 \langle |v|^3 \rangle C_2(t), \tag{18}$$

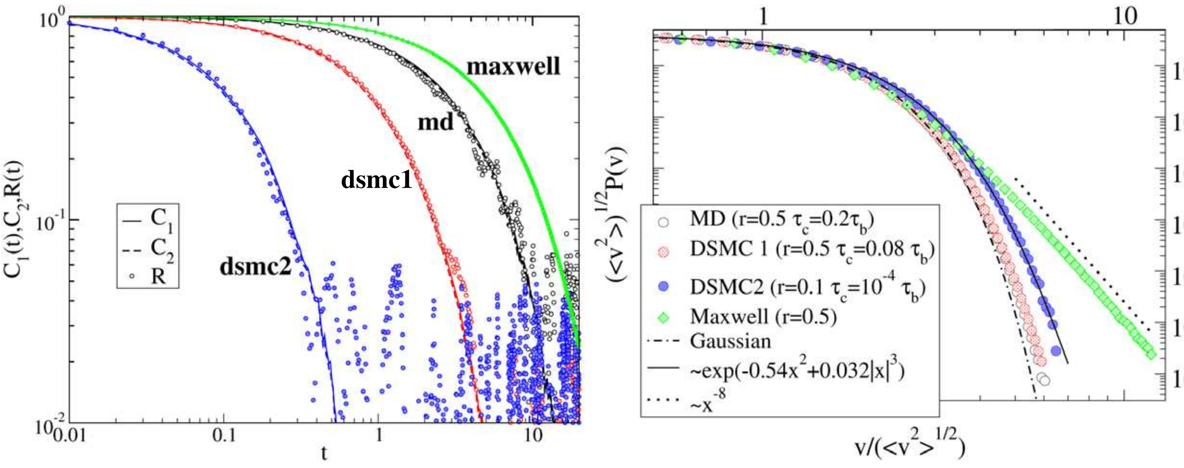

Figure 1. Left: correlation functions $C_k(t)$ and response functions $R(t)$ versus time for different models described in the text. Data for MD and DSMC come from simulations, while for the Maxwell model C_1, C_2 and R are analytical; see the appendix. Right: pdfs of the x component of the velocity (here denoted as v), for the same models. All data come from simulations with $N = 1000$. In the MD simulation the box is of size 100×100, $\tau_b = 1/\gamma = 10$ and $T_b = 1$. All other parameters are reported in the figure.

which also defines $C_2(t)$. This is an example of the discussion given in the introduction: an ansatz on the invariant measure leads to a link between the response function and some correlation functions. Note that here the ansatz is composed of two assumptions: positions and velocities of the grains are independent (this is exactly true for models (ii) and (iii)), equation (11), and a specific non-Gaussian shape of $p_v(v)$. Finally, for the inelastic Maxwell model the tail of the velocity pdf shows a power law decay with an exponent in agreement with its quasi-elastic limit $4/(1-r) = 8$ when $r = 0.5$.

In the left frame of figure 1, we have superimposed the response $\overline{\delta v(t)}/\delta v(0)$ to the time self-correlations of different orders $C_1(t)$ and $C_2(t)$ measured in the unperturbed system. From the above results, we learn that in all the considered models:

- different correlations are almost identical (we do not show $C_3(t) = \langle v(t)v^3(0)\rangle/\langle v^4\rangle$, but the result is very close);
- a very good agreement between $R(t)$ and $C_1(t)$ is observed, equivalent to a verification, within the limits of numerical precision, of the Einstein relation.

The observation that the self-correlations $C_k(t)$, at least for $k = 1, 2, 3$, are almost identical is very robust. With a precise statistics one can appreciate small differences at large times, proving that it is not an exact equivalence. Anyway, the measurement of the response function is usually very noisy, and it is not easy to have a good signal/noise ratio at such late times. Therefore, for the practical purpose of the linear combination involved in the response, these small differences are negligible and the Einstein relation is practically satisfied.

It is interesting to note that a rather similar situation is encountered when studying a different system, i.e. a gas of non-interacting particles whose velocities obey a Langevin

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equation with a non-quadratic potential:

$$\frac{\mathrm{d}v(t)}{\mathrm{d}t} = -\gamma \frac{\mathrm{d}U(v)}{\mathrm{d}v} + \sqrt{2\gamma}\eta(t), \tag{19}$$

with $U(v) = c_1 v^2 - c_2 v|v|^2 + c_3 v^4$ (with positive c_1, c_2 and c_3). A numerical inspection, not shown here, clearly indicates that $C_1(t)$, $C_2(t)$ and $C_3(t)$ are almost indistinguishable.

A simple condition can be given for the observed behaviour. In fact, a generic time correlation for $v(t)$ with a function $f[v(0)]$ can be written as

$$\langle v(t) f[v(0)]\rangle = \int \mathrm{d}v_t \int \mathrm{d}v_0 p_v(v_0)\mathcal{P}_t(v_t|v_0) v_t f(v_0) \tag{20}$$

$$= \int \mathrm{d}v_0 p_v(v_0) f(v_0)\langle v_t|v_0\rangle, \tag{21}$$

where $\mathcal{P}_t(v_t|v_0)$ is the conditional probability of observing $v(t) = v_t$ if $v(0) = v_0$ (time translation invariance is assumed) and $\langle v_t|v_0\rangle = \int \mathrm{d}v_t\, \mathcal{P}_t(v_t|v_0)v_t$ is the average of $v(t)$ conditioned to $v(0) = v_0$.

If, for some reason, $\langle v_t|v_0\rangle = g(t)q(v_0)$, with g and q two given functions, then the dependence on t results independent of the choice of the function $f(v)$, i.e. the order of the correlation. This happens in model (iii), where in spite of the non-Gaussian shape of the velocity pdf, the equivalence between $R(t)$ and $C_1(t)$ and any other correlation

$$C_f(t) = \frac{\langle v(t) f[v(0)]\rangle}{\langle v(0) f[v(0)]\rangle} = R(t) = \exp\left(-\frac{r(r+1)}{4}t\right), \tag{22}$$

with any generic function f of the initial velocity value, is exact; see the appendix for the case $d = 2$ and [19] for $d = 1$.

3. Non-homogeneous granular fluids

The factorization of the invariant phase space measure, equation (11), is no longer obvious in model (i) when density increases. Correlations between different d.o.f., that is positions and velocities of the same or of different particles, appear also in homogeneously driven granular gases, as an effect of the inelastic collisions that act similarly to an attractive potential. Such a phenomenon has been discussed for this model of a bath in [11, 34, 35] and for other homogeneous thermostats in [10, 12, 36]. In [11, 34] there was also discussed the interplay between local density and local granular temperature, which in some very dissipative cases present strong fluctuations correlated with each other. These correlations indicate a breakdown of the factorization of the invariant measure, in particular at the level of velocity with respect to position of the same particle. As a matter of fact, these effects result in a strong violation of the Einstein relation, and in general of the equivalence between $R(t)$ and $C_1(t)$.

Even in the presence of correlations, one can define and compute the marginal probability density function of the component x of the velocity of one particle i, projecting the phase space measure $\rho(\{\mathbf{v}_i, \mathbf{x}_i\})$:

$$f_i(v) = \int \prod_{k=1}^{N} \mathrm{d}\mathbf{x}_i \prod_{k=1, k\neq i}^{N} \mathrm{d}\mathbf{v}_k\, \mathrm{d}v_i^y \rho(\{\mathbf{v}, \mathbf{x}\}). \tag{23}$$

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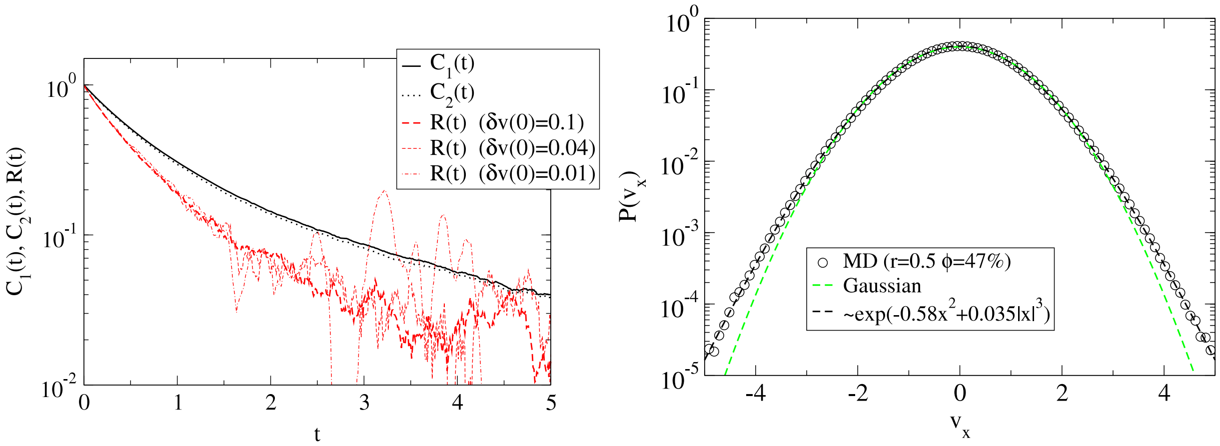

Figure 2. Left: correlation functions $C_k(t)$ and response functions $R(t)$ versus time for a dense MD simulation: the response function is reported for different values of the perturbation $\delta v(0)$. Right: pdf of the x velocity component. The system has $N = 1000$, box of size 41×41, $\tau_{\mathrm{b}} = 1/\gamma = 10$. In the simulation the mean free time between collisions is measured to be $\tau_{\mathrm{c}} = 0.03\tau_{\mathrm{b}}$.

However this function does not necessarily have a role in the response function. For example, perturbing the x component of the velocity of the ith particle and measuring the response of the same component, one obtains

$$R(t) = -\left\langle v_i^x(t) \frac{\partial \ln \rho(\{\mathbf{v}, \mathbf{x}\})}{\partial v_i^x}\bigg|_{t=0} \right\rangle \neq -\left\langle v_i^x(t) \frac{\partial \ln f_i(v_i^x)}{\partial v_i^x}\bigg|_{t=0} \right\rangle. \quad (24)$$

This is exactly what happens in model (i) when density is increased. In figure 2, left frame, the correlation functions $C_1(t)$ and $C_2(t)$ are shown, together with the response function measured with different values of the perturbation $\delta v(0)$. The very good agreement between different response functions guarantees that the system is indeed linearly perturbed. At the same time, the different correlation functions $C_k(t)$ are very close, reproducing the phenomenology already observed in the previous dilute cases, with the difference that the time dependence is not exponential but slower, closer to a stretched exponential $\sim \exp(-(t/\tau)^\alpha)$ with $\alpha < 1$. Finally, looking at the velocity pdf of the gas, the previously proposed exponential of a cubic polynomial, equation (16) with a negligible c_3 coefficient, is found to perfectly fit the numerical results. Therefore, if the correlations among the different d.o.f. are neglected, using equation (17) and the proportionality of the functions $C_k(t)$, a verification of the Einstein formula $R(t) \simeq C_1(t)$ is still expected. The results displayed in figure 2, left frame, demonstrate that this is not the case: the hypothesis of weak correlations among different d.o.f. must be dropped and the correct formula for the response is equation (24). Unfortunately it is not very easy to use such a relation.

The degree of violation of the Einstein formula increases with the volume fraction ϕ and the inelasticity $1-r$, as shown in figure 3, where we have reported the ratio $R(t)/C_1(t)$ as a function of time. This observation is consistent with the above argument: correlations among different d.o.f. increase when the probability of repeated contacts (the so-called 'ring collisions') is enhanced, and this happens when the excluded volume and/or the post-

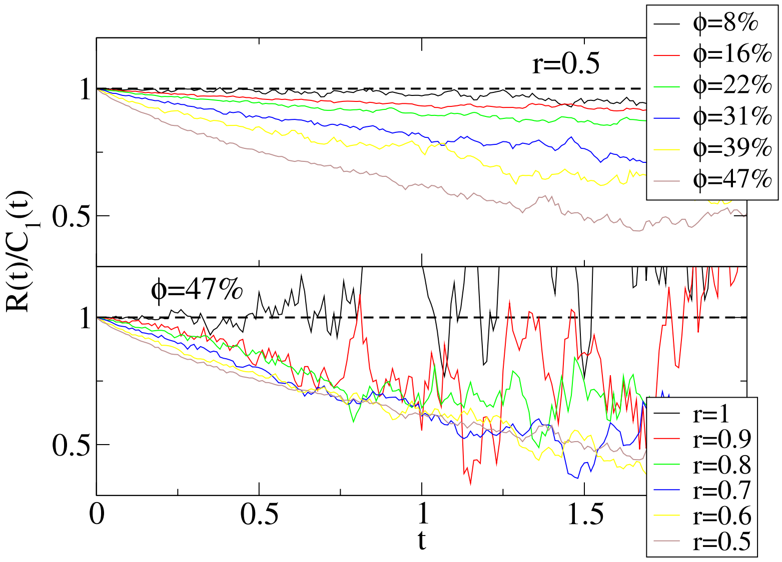

Figure 3. Ratio between the response function $R(t)$ and the normalized velocity self-correlation $C_1(t)$. The ratio is 1 when the Einstein relation is satisfied. All the results come from MD simulations with $N = 1000$ particles, $T_{\rm b} = 1$ and $\tau_{\rm b} = 1/\gamma = 10$. Different values of the covered fraction and of the restitution coefficient are used, as shown in the figure.

collisional relative velocity are reduced. In the elastic case, $r = 1$, no violation is observed. A direct test of the existence of non-trivial correlations in the system is given in figure 4. Each point in this figure represents the value of $C_{vN} = (\langle v_i^2 N_i\rangle - \langle v_i^2\rangle\langle N_i\rangle)/(\langle v_i^2\rangle\langle N_i\rangle)$ for a given particle i, with N_i the number of particles in a squared box centred at $\mathbf{x}_i$ and of size $L/15$, measured on a long trajectory of the unperturbed system. We observe that C_{vN} increases together with the volume fraction ϕ.

3.1. A Langevin model with two correlated variables

In order to show in a clear way the role of correlations, we discuss now a simple model with only two variables:

$$\frac{{\rm d}x(t)}{{\rm d}t} = m_{11}x(t) + m_{12}v(t) + \sigma_{11}\eta_1(t) + \sigma_{12}\eta_2(t) \tag{25}$$

$$\frac{{\rm d}v(t)}{{\rm d}t} = m_{12}x(t) + m_{22}v(t) + \sigma_{21}\eta_1(t) + \sigma_{22}\eta_2(t). \tag{26}$$

If the matrices $\hat{m}$ and $\hat{\sigma}$ are diagonal, the two variables are independent. Provided that the symmetric matrix $\hat{m}$ has negative eigenvalues and $\det\hat{\sigma} \neq 0$, the pdf of (x, v) relaxes toward a bi-variate Gaussian function. Instead of discussing the general form, we consider the case whose invariant joint pdf is

$$\rho(x, v) \propto \exp\left(-\frac{x^2}{2} - \frac{v^2}{2} + \frac{xv}{2}\right). \tag{27}$$

Of course the marginal pdf of each single variable is a Gaussian. Neglecting the correlation between x and v, the response of v to a perturbation of itself would again be expected to

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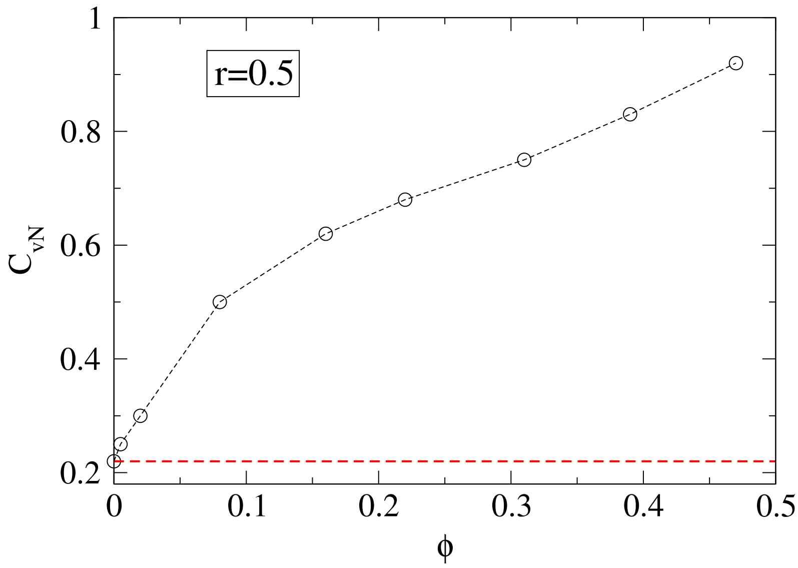

Figure 4. Correlation between the square of the particle x velocity and local density $C_{vN} = (\langle v_i^2 N_i \rangle - \langle v_i^2 \rangle \langle N_i \rangle)/(\langle v_i^2 \rangle \langle N_i \rangle)$ for different values of the volume fraction, in MD simulations with $N = 1000$, $T_\mathrm{b} = 1$ and $\tau_\mathrm{b} = 10$. In the dilute limit $\phi \to 0$, $C_{vN} \to C^*_{vN}$, which is different from zero because of the total finite number of particles. The red dashed line shows the estimate of C^*_{vN} obtained by throwing N random velocities, extracted from a Gaussian distribution, into random boxes of the same size as is used in the MD, and repeating the measurement over many independent realizations.

be equal to $C_1(t) = \langle v(t)v(0)\rangle / \langle v^2 \rangle$. In contrast, the correct response is given using the full formula (8) applied to the joint pdf (27). The result is

$$R(t) = \langle v(t)v(0)\rangle - \tfrac{1}{2}\langle v(t)x(0)\rangle. \tag{28}$$

The difference between the Einstein formula and the correct response is shown in figure 5 for a choice of the matrix $\hat{m}$.

4. Conclusions

In this paper we have reported the analysis of linear response in different models of driven granular gases, which have the property of rapidly reaching a statistically stationary state. The response function is directly related to the global invariant measure in the phase space, which is unknown for such non-equilibrium systems. When positions and velocities of the particles are not correlated, the response to a perturbation of the velocity of a single particle is expected to depend on the singlet velocity pdf, which can be close to or far from a Gaussian, depending on the model and on physical parameters. Nevertheless, the existence of a unique timescale that characterizes all possible correlation functions makes the exact form of the velocity pdf irrelevant for the response function: the latter is, in practice, always indistinguishable from the normalized velocity self-correlation $\langle v(t)v(0)\rangle / \langle v^2 \rangle$. This is equivalent to saying that the Einstein relation is satisfied for all heated granular systems where correlations among particles are weak. On the other hand,

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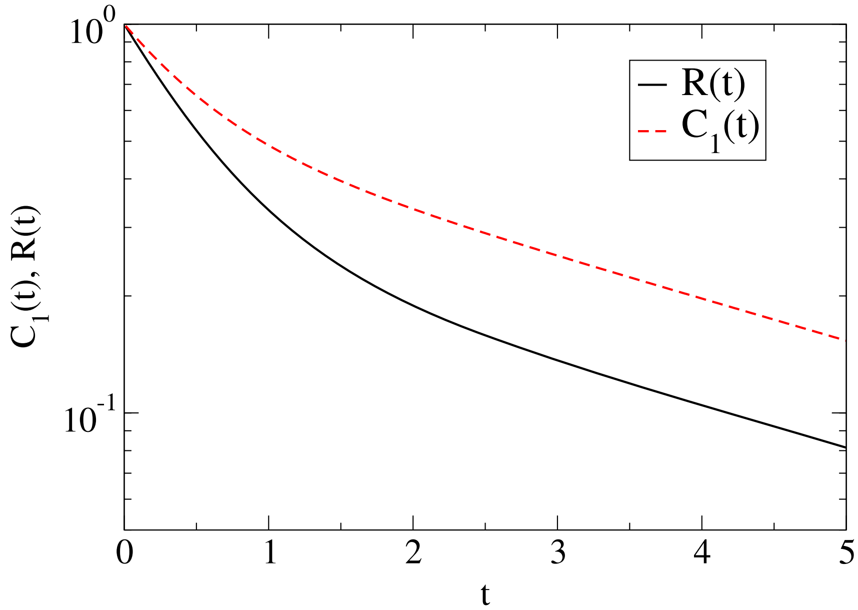

Figure 5. Response $R(t)$ and velocity correlation $C_1(t)$ in the simple Langevin model with correlated variables discussed in equation (25), with parameters $m_{11} = -1.1$, $m_{12} = 0.8$, $m_{22} = -1$.

when the excluded volume and energy dissipation occurring in collisions are increased, non-trivial correlations appear among positions and velocities of particles. The global invariant measure cannot be factorized any longer and the response function depends on it, i.e. on the specific parameters of the model. As a consequence, the Einstein relation is no longer satisfied and the response function is not trivially predictable. In all simulations, the decay of $R(t)$ is always faster than that of $C_1(t)$: this is equivalent to stating for the mobility that

$$\mu = \frac{1}{m}\int_0^\infty \mathrm{d}t\, R(t) < \frac{D}{T_\mathrm{g}}. \tag{29}$$

It should be noted that for the gas of inelastic hard spheres with constant kinetic energy (obtained with the 'Gaussian thermostat'), even when homogeneous, a small but appreciable (∼5%) discrepancy between mobility and diffusion is expected, as discussed in [21].

Inspired by a recent work which reported violations of the Einstein relation in a non-equilibrium model [37], we now conjecture an effective spatial dependence of the pdf of the velocity component for a particle at position $\mathbf{x}$ at time t of the form

$$p_v(v, \mathbf{x}, t) \sim \exp\left\{-\frac{[v - u(\mathbf{x}, t)]^2}{2T_\mathrm{g}}\right\}, \tag{30}$$

with $u(\mathbf{x}, t)$ a local velocity average, defined on a small cell of diameter L_box centred at the particle. Such a hypothesis is motivated by the fact that, at high density or inelasticity, spatially structured velocity fluctuations appear in the system for some time, even in the presence of external noise [12, 36]. Following relation (8) we propose a formula for the response function for a velocity perturbation:

$$R(t) = C_s = \frac{1}{T_\mathrm{g}}\langle v(t)\{v(0) - u[x(0)]\}\rangle. \tag{31}$$

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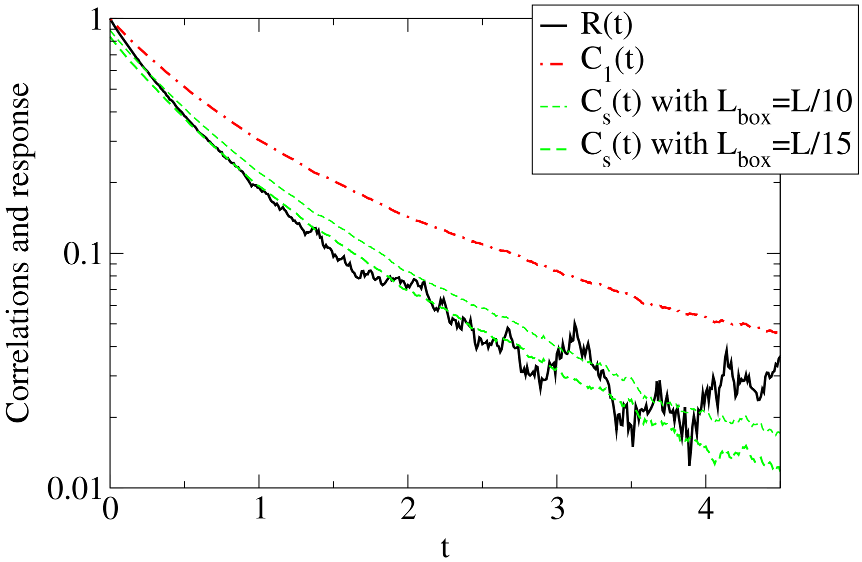

Figure 6. Response $R(t)$ and different correlation functions for the same MD simulation discussed in figure 2. The normalized velocity self-correlations $C_1(t)$, as well as the correlation $C_s(t)$ defined in equation (31), for different values of the coarse graining radius $L_{\rm box}$ are reported.

Figure 6 shows that for small values of the coarse graining diameter $L_{\rm box}$ (but still large enough to include 5–10 particles) relation (31) is fairly well verified. Note however that the proposed form (30) cannot be exact; a spatial dependence of $T_{\rm g}$ should also be included. Furthermore, it is clear that $\langle u(\mathbf{x})\rangle = 0$ for any point $\mathbf{x}$, i.e. the local velocity field $u(\mathbf{x})$ fluctuates in time. Thus, the above conjecture implies that the characteristic time of variation of these fluctuations is larger than the characteristic time of response of a particle: the particle feels, during its response dynamics, the 'local equilibrium' average $u(\mathbf{x})$. Further investigations are in progress to refine this promising argument. Moreover, in order to evaluate the role of the violations of molecular chaos due to excluded volume effects, simulations in the Enskog approximation (using a non-homogeneous DSMC algorithm) are included in our ongoing investigation.

Acknowledgments

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Appendix. The inelastic Maxwell model

The dynamics of a particle in the inelastic Maxwell model is described by the following stochastic process:

$$\mathbf{v}(t+\Delta t) - \mathbf{v}(t) = \begin{cases} \lambda \mathbf{v}(t)\Delta t & \text{(with prob. } 1-\Delta t) \\ \lambda \mathbf{v}(t)\Delta t + \Delta \mathbf{v}_{\rm col}(\mathbf{v},\mathbf{u},\boldsymbol{\sigma}) & \text{(with prob. } \Delta t) \end{cases} \tag{A.1}$$

where $\Delta \mathbf{v}_{\rm col}(\mathbf{v},\mathbf{u},\boldsymbol{\sigma})$ is the effect of an inelastic collision and has been defined in equation (15), and $\mathbf{u}$ is the velocity of the collision partner.

Let us define the two-times covariance matrix $A_{\mu\nu}(t_1,t_2) = \langle v_\mu(t_1)v_\nu(t_2)\rangle$, with $\mu,\nu \in \{x,y\}$. Using the evolution law of the system, equation (A.1), one can calculate

$$\frac{\partial A_{\mu\nu}}{\partial t_2} = \lim_{\Delta t_2\to 0}\left\langle v_\mu(t_1)\frac{v_\nu(t_2+\Delta t_2)-v_\nu(t_2)}{\Delta t_2}\right\rangle \tag{A.2}$$

$$= \langle v_\mu(t_1)[\lambda v_\nu(t_2)+\Delta v_{\mathrm{col},\nu}]\rangle \tag{A.3}$$

$$= \lambda\langle v_\mu(t_1)v_\nu(t_2)\rangle - \frac{1+r}{2}\langle v_\mu(t_1)\sigma_\nu[[\mathbf{v}(t_2)-\mathbf{u}(t_2)]\cdot\boldsymbol{\sigma}]\rangle \tag{A.4}$$

$$= \lambda\langle v_\mu(t_1)v_\nu(t_2)\rangle \tag{A.5}$$

$$-\frac{1+r}{2}\langle v_\mu(t_1)[v_x(t_2)\sigma_x - u_x(t_2)\sigma_x + v_y(t_2)\sigma_y - u_y(t_2)\sigma_y]\sigma_\nu\rangle \tag{A.6}$$

$$= \lambda\langle v_\mu(t_1)v_\nu(t_2)\rangle - \frac{1+r}{2}[\langle v_\mu(t_1)v_x(t_2)\sigma_x\sigma_\nu\rangle + \langle v_\mu(t_1)v_y(t_2)\sigma_y\sigma_\nu\rangle \tag{A.7}$$

$$-\ \langle v_\mu(t_1)u_x\sigma_x\sigma_\nu\rangle - \langle v_\mu(t_1)u_y\sigma_y\sigma_\nu\rangle] \tag{A.8}$$

which, assuming absence of correlation between pre-collisional velocities of different particles and between them and the impact vector $\boldsymbol{\sigma}$, gives

$$\frac{\partial A_{\mu\nu}}{\partial t_2} \tag{A.9}$$

$$= \lambda\langle v_\mu(t_1)v_\nu(t_2)\rangle - \frac{1+r}{2}[\langle v_\mu(t_1)v_x(t_2)\rangle\langle\sigma_x\sigma_\nu\rangle + \langle v_\mu(t_1)v_y(t_2)\rangle\langle\sigma_y\sigma_\nu\rangle] \tag{A.10}$$

$$= \kappa A_{\mu\nu} \tag{A.11}$$

where $\kappa = (\lambda - (1+r)/4) = -r(r+1)/4$, and we have used the fact that $\langle\sigma_\mu\sigma_\nu\rangle = (1/2)\delta_{\mu\nu}$. Since the system is time translation invariant, we have that $A_{\mu\mu}(t) = A_{\mu\nu}(0)\exp(\kappa(t_2-t_1))$.

Now, one can perturb at a certain time the velocity $\mathbf{v}$ of a unique particle, with such a small perturbation that it does not modify the rest of the system. Starting from (A.1), one easily computes the average response to such a perturbation:

$$\frac{\mathrm{d}\langle\mathbf{v}\rangle}{\mathrm{d}t} = \lambda\langle\mathbf{v}\rangle - \frac{1+r}{2}\langle\mathbf{F}(\mathbf{v},\mathbf{u},\hat{\boldsymbol{\sigma}})\rangle = \left(\lambda - \frac{1+r}{4}\right)\langle\mathbf{v}\rangle, \tag{A.12}$$

where $\mathbf{F}(\mathbf{v},\mathbf{u},\hat{\boldsymbol{\sigma}}) = (v_x\sigma_x^2 - u_x\sigma_x^2 + v_y\sigma_x\sigma_y - u_y\sigma_y\sigma_x, v_x\sigma_x\sigma_y - u_x\sigma_x\sigma_y + v_y\sigma_y^2 - u_y\sigma_y^2)$ and we used the fact that $\langle\mathbf{u}\rangle = 0$. The result is that the average response decays as the velocity–velocity correlation.

A more general result can be also obtained, starting from the stochastic evolution equation (A.1), which can be rephrased as

$$\mathbf{v}(t+\Delta t) = \hat{K}(t)\mathbf{v}(t) + \hat{J}(t) \tag{A.13}$$

where $\hat{K}(t)$ and $\hat{J}(t)$ are two uncorrelated stochastic (two-dimensional) matrices, with $\langle K_{\mu\nu}(t)\rangle \neq 0$ and $\langle J_{ij}(t)\rangle = 0$. Starting at time 0 with $\mathbf{v}(0) = \mathbf{v}_0$ and iterating equation (A.13), one finds that $\langle\mathbf{v}(t)|\mathbf{v}_0\rangle = \hat{L}^t\mathbf{v}_0$ where $L_{\mu\nu} = \langle K_{\mu\nu}\rangle$. From relation (20) (generalized to two dimensions), the proportionality of all the correlation functions follows.

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