Green-Kubo relations for granular fluids

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A formulation of response theory for the dynamics of moderately dense granular gases is presented. Some implications concerning the derivation of constitutive relations for granular gases at finite densities are presented.

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In spite of much recent progress in describing the kinetics and hydrodynamics of granular gases, the solution of this problem is far from being complete. For instance, the fact that the typical expansion parameter in the Chapman-Enskog expansion of the Boltzmann equation for granular gases is not small [1] requires that the Burnett and super-Burnett contributions [2] be included in the hydrodynamic description [3], the former being the source of the significant normal stress differences in granular gases [3]. As the Burnett and super-Burnett equations are ill-posed [4], they can be directly used only for the study of steady states, else one needs to "resum" the Chapman-Enskog series [4]. Furthermore, in employing the Chapman-Enskog perturbative expansion, the zeroth-order single-particle distribution function is taken to be Gaussian [3] (when inelasticity is accounted for perturbatively) or that corresponding to an unforced decaying flow [5]. As recent experiments have shown [6] the actual distribution functions (in vibrated systems) may be strongly non-Gaussian and not just small perturbations of a Maxwellian. Encouragingly, distribution functions that do correspond to experimental findings have been derived from a model of randomly forced grains [7].

The above and other problems arise already in the domain of dilute granular gases. It is no less important to study the dynamics of moderately dense granular fluids. In this domain only semiphenomenological approaches, mostly based on the application of the Enskog-Boltzmann equation [8] have been implemented. As these approaches fail to account for correlated collisions, which are of importance in nondilute gases, it is unclear whether they produce reliable equations of motion. It is the goal of this article to present a formulation that enables a systematic approach to the description of nondilute granular fluids. To this end we present a modification of response theory [9], which has proven to be a powerful tool in the realm of molecular gases.

Standard response theory [9] is based on the following thought experiment: a system is prepared in the 'infinite' past in a state of equilibrium and a set of adiabatically increasing forces is applied to it up to a time t=0, at which these forces are switched off. The hydrodynamic equations are then deduced from the dynamics of decay to equilibrium of the state at t=0 (assuming Onsager's regression hypothesis). A perturbation theory to second order in the forces begets the Navier-Stokes equations. This approach is not directly appli-

cable to granular systems as their energy decays at a finite rate due to the dissipative nature of the grain interactions. However, a variant of the above thought experiment [10], in which the system is prepared at t=0 in a state of local equilibrium, seems to be appropriate to granular systems. It is the latter approach that serves as the basis for the formulation presented below. A modification of this approach is shown to yield Green-Kubo expressions for the transport coefficients corresponding to granular fluids. The formulation presented below is restricted, for sake of simplicity, to the case of a monodisperse collection of spheres [in three dimension (3D)] whose collisions are characterized by a fixed coefficient of normal restitution.

A key role in the formulation presented below is played by the (idealized) homogeneous and isotropic unforced granular state. As is well known, this state is unstable [11,12]. However, it has proven useful as a zeroth order in a perturbative analysis of the Boltzmann equation corresponding to the above model of a granular medium [5] and it's use as a replacement for the equilibrium state in the response formulation does not create any difficulties. It is rather obvious that the dynamics of this state amounts to a decrease in energy alone. It follows that the full many-body distribution function corresponding to this state, which we denote by ρ_H , satisfies the following scaling relation:

$$\rho_H(\boldsymbol{\chi},t) = \frac{1}{v_0^{3N}(t)} \tilde{\rho}_H(\{\mathbf{r}_i,\mathbf{c}_i\}),\tag{1}$$

where $\mathbf{v}_i(t)$ is the velocity of particle *i* at time *t*, $\mathbf{c}_i = \mathbf{v}_i/v_0(t)$, $v_0(t)$ is the thermal velocity characterizing the homogeneous ensemble (the square root of the granular temperature, *T*) and χ represents the phase space of the system. Since $\tilde{\rho}_H(\{\mathbf{r}_i, \mathbf{c}_i\})$ is time independent, averages of dynamical variables which depend only on $\{\mathbf{r}_i, \mathbf{c}_i\}$, with respect to ρ_H , are time independent. It is convenient to measure time in this system by the (average) accumulated number of collisions, τ . It is easy to see that, with this definition of 'time', unequal time correlation functions of properly rescaled fields (see below) in the ensemble defined by ρ_H depend on differences of the respective values of τ and in this sense the correlations functions are time translational invariant (when velocity dependent coefficients of restitution are allowed for, the above statements have to be modified). The time dependent

3241

dence of the thermal velocity, $v_0(t)$, in the above homogeneous ensemble, is determined by the energy dissipation rate, which depends on the number density, n, and T(t). Since, by assumption, the coefficient of normal restitution, e, is constant, the decay rate of T must be (as is well known) proportional to the collision frequency $\omega(t) \propto \sqrt{T(t)}$. One can thus define a dimensionless cooling rate, γ_H , by the relation $\dot{T} = -2\gamma_H\omega(t)T(t)$, which describes the time dependence of the granular temperature of the homogeneous system. In general, γ_H depends on n. In the dilute limit, in 3D, and to first order in the degree of inelasticity, $\epsilon = 1 - e^2$, it is given by $\gamma_0 = \epsilon/6$. The accumulated average number of collisions per particle, τ , satisfies: $d\tau = \omega(t)dt$. The thermal velocity decays exponentially in $\tau: v_0(t) = v_0(0) \exp[-\gamma_H \tau]$.

Let $\{A\}$ denote the set of macrovariables (hydrodynamic fields) for which one wishes to obtain equations of motion. Below we take this set to consist of the number density, $N(\mathbf{r},t)$, the momentum density, $\mathbf{P}(\mathbf{r},t)$, and the energy density, $E(\mathbf{r},t)$ (though the latter is not, strictly speaking, "slow" because energy is "lost" in collisions). The equations of motion satisfied by the set of macrofields can be written as follows: $\partial_t \mathbf{A}(\mathbf{r},t) = -\nabla \cdot \mathbf{J}(\mathbf{r},t) - \Gamma(\mathbf{r},t)$, where $\mathbf{A}(\mathbf{r},t) \equiv \mathbf{A}(\mathbf{r},\chi(t))$. With the above choice of fields the only nonzero component of Γ is that corresponding to the energy equation; it is denoted by Γ^{e} . The energy density flux is denoted by \mathbf{J}^{e} , the (tensorial) momentum density flux by \mathbf{J}^{p} and the number density flux, $n\mathbf{v}$, is denoted by \mathbf{J}^n . It is convenient to define rescaled fields as follows: { $\tilde{A}^{a}(\mathbf{r},\tau)$ $\equiv A^{a}(\mathbf{r},t)/v_{0}^{q^{a}}(t)$, where the powers of v_{0} are given by: $q^n = 0, q^p = 1$ and $q^e = 2$, corresponding to the number density, momentum density and energy density fields, respectively. The corresponding fluxes and dissipation rates are rescaled according to: $\tilde{\mathbf{J}}^{a}(\mathbf{r},\tau) = \mathbf{J}^{a}(\mathbf{r},t)/v_{0}^{q^{a}+1}(t)$ and $\tilde{\Gamma}^{a}(\mathbf{r},\tau)$ $= \Gamma^a(\mathbf{r},t) / v_0^{q^a+1}(t).$

Following Ref. [10], assume the following initial distribution function, at t=0:

$$\rho_{NE}(\chi, t=0) = \frac{\rho_H(\chi, 0) \exp(\mathbf{\tilde{A}} * \mathbf{F})}{\langle \exp(\mathbf{\tilde{A}} * \mathbf{F}) \rangle_H},$$
(2)

where ρ_H is the above defined distribution function considered in a grand canonical ensemble, and the asterisk represents integration over space and summation over discrete indices. It is convenient to chose (without loss of generality) a frame of reference in which the total momentum vanishes. As in Ref. [10], the 'forces' or chemical potentials $\{\mathbf{F}\}$ are chosen such that the averages of the set $\{A\}$ at a predetermined time, t, (which should be large with respect to the mean-free time) have specified values $\mathbf{a}(t) \equiv \langle \mathbf{A}(t) \rangle_{NE}$ and the role of the theory is to determine the time derivatives of these entities (or the values of the fluxes and the sink term) at time t. The basic assumption of this formulation is that if t is far larger than the mean-free time the distribution function at time t is determined by the above (average) values of the fields at time t and not by the precise form of the initial condition Eq. (2) (this issue is explained in some detail in Ref. [10] and Refs. therein). The nonequilibrium average of any dynamical variable, B(t), is given by: $\langle B(t) \rangle_{NE}$ = Tr[$B(t)\rho_{NE}(\chi,0)$]. Following this definition and Eq. (2) the nonequilibrium average of any dynamical variable can be expanded in powers of the forces {**F**}, the "prefactors" being cumulants of the fluctuations of the fields { $\widetilde{\mathbf{A}}$ }. Upon substituting the fields $\widetilde{\mathbf{A}}(\tau)$ for B(t) in the above expression one obtains an implicit equation for the chemical potentials, which can be formally inverted (or practically, by employing perturbation theory) to express the chemical potentials {**F**} in terms of cumulants of the fields (for details see Ref. [10]). The resulting expression, to first order in the fluctuations, reads: $\mathbf{F}(\{\delta \widetilde{\mathbf{a}}(\tau)\}) = \langle \delta \widetilde{\mathbf{A}}(\tau) \delta \widetilde{\mathbf{A}} \rangle_{H}^{-1} * \delta \widetilde{\mathbf{a}}(\tau) + \mathcal{O}(\delta \widetilde{\mathbf{a}}^{2}(\tau)),$ where $\delta \widetilde{\mathbf{A}}(\tau) \equiv \widetilde{\mathbf{A}}(\tau) - \langle \widetilde{\mathbf{A}} \rangle_{H}$ and $\delta \widetilde{\mathbf{a}} \equiv \langle \delta \widetilde{\mathbf{A}} \rangle_{NE}$. As this procedure determines ρ_{NE} , one can use it to expand the nonequilibrium average of any function of the phase variables of the system, e.g.,

$$\langle B(t) \rangle_{NE} = \langle B(t) \rangle_{H} + \langle \delta B(t) \, \delta \widetilde{\mathbf{A}} \rangle_{H} * \langle \delta \widetilde{\mathbf{A}}(\tau) \, \delta \widetilde{\mathbf{A}} \rangle_{H}^{-1} * \delta \widetilde{\mathbf{a}}(\tau) + \mathcal{O}(\delta \widetilde{\mathbf{a}}^{2}(\tau)).$$
(3)

We shall not dwell here on the nonlinear terms as their expressions are rather lengthy. We wish to stress though that the formulation presented above is valid, in principle, "to all orders in \mathbf{F} ," cf. also Ref. [10]. The next step in carrying out the response formulation towards the achievement of a hydrodynamic description is a gradient expansion. A standard procedure [9,10] yields from Eq. (3), to first order in the gradient expansion:

$$\langle B(\mathbf{r},t) \rangle_{NE} = \langle B(r,t) \rangle_{H} + \langle \delta B(\mathbf{r},t) \delta \widetilde{\mathbf{A}}(\mathbf{r}_{2}) \rangle_{H} * \\ \times \langle \delta \widetilde{\mathbf{A}}(\mathbf{r}_{2},\tau) \delta \widetilde{\mathbf{A}}(\mathbf{r}_{1}) \rangle_{H}^{-1} * (\mathbf{r}_{1}-\mathbf{r}) \cdot \nabla \widetilde{\mathbf{a}}(\mathbf{r},\tau),$$

$$(4)$$

where the spatial dependence is now explicitly spelled out. Using the spatial Fourier transform, $B(\mathbf{k},t) = \int d\mathbf{r} \exp(-i\mathbf{k}\cdot\mathbf{r})B(\mathbf{r},t)$, the right hand side of Eq. (4) transforms to

$$\langle B(\mathbf{r},t) \rangle_{NE} = \langle B(\mathbf{r},t) \rangle_{H} + v_{0}^{q^{b}}(t) \frac{\partial}{\partial i k_{\alpha}} \\ \times \widetilde{\mathbf{M}}(\mathbf{k},\tau) \bigg|_{k=0} \cdot \nabla_{\alpha} \widetilde{\mathbf{a}}(\mathbf{r},\tau),$$
 (5)

where

$$\widetilde{\mathbf{M}}(\mathbf{k},\tau) \equiv \langle \, \delta \widetilde{B}(\mathbf{k},\tau) \, \delta \widetilde{\mathbf{A}}(-\mathbf{k}) \rangle_{H} \cdot \langle \, \delta \widetilde{\mathbf{A}}(\mathbf{k},\tau) \, \delta \widetilde{\mathbf{A}}(-\mathbf{k}) \rangle_{H}^{-1},$$

which corresponds to the hydrodynamic matrix (in scaled variables) when $\tilde{B}(\tau) \equiv B(t)/v_0^{q^b}(t)$ is substituted for by $\tilde{\mathbf{J}}$ or $\tilde{\Gamma}^e$. Following standard practice [9,10], it is useful to write

$$\widetilde{\mathbf{M}}(\mathbf{k},\tau) = \widetilde{\mathbf{M}}(\mathbf{k},0) + \int_{0}^{\tau} d\tau' \widetilde{\mathbf{M}}_{\tau}(\mathbf{k},\tau'), \qquad (6)$$

where the subscript τ denotes a derivative with respect to τ . The isotropy and homogeneity of the homogeneous en-

$$\begin{split} \widetilde{\mathbf{M}}_{\tau}(\mathbf{k},\tau) &= \langle \widetilde{B}_{\tau}(\mathbf{k},\tau) \, \delta \widetilde{\mathbf{A}}(-\mathbf{k}) \rangle_{H} \cdot \langle \delta \widetilde{\mathbf{A}}(\mathbf{k},\tau) \, \delta \widetilde{\mathbf{A}}(-\mathbf{k}) \rangle_{H}^{-1} \\ &- \langle \delta \widetilde{B}(\mathbf{k},\tau) \, \delta \widetilde{\mathbf{A}}(-\mathbf{k}) \rangle_{H} \cdot \langle \delta \widetilde{\mathbf{A}}(-\mathbf{k}) \rangle_{H} \cdot \langle \delta \widetilde{\mathbf{A}}(\mathbf{k},\tau) \\ &\times \delta \widetilde{\mathbf{A}}(-\mathbf{k}) \rangle_{H}^{-1} \cdot \langle \widetilde{\mathbf{A}}_{\tau}(\mathbf{k},\tau) \\ &\times \delta \widetilde{\mathbf{A}}(-\mathbf{k}) \rangle_{H} \cdot \langle \delta \widetilde{\mathbf{A}}(\mathbf{k},\tau) \, \delta \widetilde{\mathbf{A}}(-\mathbf{k}) \rangle_{H}^{-1}. \end{split}$$
(7)

Next, the time translational invariance of time correlations of the scaled fields in the homogeneous scaling ensemble, i.e., $(d/d\tau')\langle \tilde{B}(\tau+\tau')\tilde{A}(\tau')\rangle_{H}=0$, makes possible a "switching" of the τ derivative in correlations: $\langle \tilde{B}_{\tau}(\tau)\tilde{A}(0)\rangle$ $= -\langle \tilde{B}(\tau)\tilde{A}_{\tau}(0)\rangle$. Using this property, one obtains from Eq. (7):

$$\partial_{\tau} \widetilde{\mathbf{M}}(\mathbf{k}, \tau) = - \langle \widetilde{B}_{D}(\mathbf{k}, \tau) \widetilde{\mathbf{A}}_{\tau}(-\mathbf{k}) \rangle_{H} \cdot \langle \delta \widetilde{\mathbf{A}}(\mathbf{k}, \tau) \delta \widetilde{\mathbf{A}}(-\mathbf{k}) \rangle_{H}^{-1},$$
(8)

where the dissipative part, $\tilde{B}_D(\tau)$, is defined (as usual) as its projection orthogonal to the relevant fluctuations $\delta \tilde{\mathbf{A}}(\tau)$, i.e., $\tilde{B}_D(\tau) \equiv \tilde{B}(\tau) - \langle \tilde{B}(\tau) \delta \tilde{\mathbf{A}} \rangle_H^* \langle \delta \tilde{\mathbf{A}}(\tau) \delta \tilde{\mathbf{A}} \rangle_H^{-1} * \delta \tilde{\mathbf{A}}(\tau)$. At this stage it is convenient to express the general equations of motion for the rescaled fields in terms or the rescaled fluxes. A straightforward calculation yields:

$$\begin{split} \widetilde{A}_{\tau}(\mathbf{k},\tau) &= -l_0 [i\mathbf{k} \cdot \widetilde{\mathbf{J}}^a(\mathbf{k},\tau) + \widetilde{\Gamma}^a(\mathbf{k},\tau)] + q^a \gamma_H \widetilde{A}(\mathbf{k},\tau) \\ &= -l_0 [i\mathbf{k} \cdot \widetilde{\mathbf{J}}^a(\mathbf{k},\tau) + \delta \widetilde{\Gamma}^a(\mathbf{k},\tau)] + q^a \gamma_H \delta \widetilde{A}(\mathbf{k},\tau), \end{split}$$
(9)

where $\delta \tilde{\Gamma}^a \equiv \tilde{\Gamma}^a - \langle \tilde{\Gamma}^a \rangle_H$. The length $l_0 \equiv v_0(t)/\omega(t)$ is the (time independent) mean free path (in the homogeneous ensemble). Note that the right hand side of Eq. (9) vanishes upon averaging over the homogeneous ensemble, as it should since $\langle \tilde{A}(\mathbf{k},\tau) \rangle_H$ is time independent (actually, it is also zero except for $\mathbf{k}=\mathbf{0}$). Using Eq. (9) together with $\langle \tilde{B}_D(\mathbf{k},\tau) \delta \tilde{A} (-\mathbf{k}) \rangle_H = 0$, we obtain

$$\langle \widetilde{B}_D(\mathbf{k},\tau) \widetilde{\mathbf{A}}_{\tau}(-\mathbf{k}) \rangle_H$$

= $-l_0 \langle \widetilde{B}_D(\mathbf{k},\tau) [-i\mathbf{k} \cdot \widetilde{\mathbf{J}}(-\mathbf{k},\tau) + \delta \widetilde{\mathbf{\Gamma}}(-\mathbf{k},\tau)] \rangle_H,$ (10)

which, together with Eqs. (8) and (6), yields for Eq. (5)

$$\begin{split} \langle B(\mathbf{r},t) \rangle_{NE} &= \langle B(\mathbf{r},t) \rangle_{H} - l_{0} v_{0}^{q^{b}}(t) \\ &\times \int_{0}^{\infty} d\tau \langle \widetilde{B}_{D}(\mathbf{0},\tau) \widetilde{\mathbf{J}}_{\alpha}(\mathbf{0}) \rangle_{H} \cdot \langle \delta \widetilde{\mathbf{A}}(0,\tau) \\ &\times \delta \widetilde{\mathbf{A}}(\mathbf{0}) \rangle_{H}^{-1} \cdot \nabla_{\alpha} \widetilde{\mathbf{a}}(\mathbf{r},\tau) + l_{0} v_{0}^{q^{b}}(t) \\ &\times \int_{0}^{\infty} d\tau \frac{\partial}{\partial i k_{\alpha}} (\langle \widetilde{B}_{D}(\mathbf{k},\tau) \delta \widetilde{\mathbf{\Gamma}}(-\mathbf{k}) \rangle_{H} \langle \delta \widetilde{\mathbf{A}}(\mathbf{k},\tau) \\ &\times \delta \widetilde{\mathbf{A}}(-\mathbf{k}) \rangle_{H}^{-1}) |_{k=0} \cdot \nabla_{\alpha} \widetilde{\mathbf{a}}(\mathbf{r},\tau), \end{split}$$

where the integrations over τ have been extended to infinity, anticipating rapid convergence of the corresponding integrals. In molecular fluids, the last term in Eq. (11), involving the dissipation rate, vanishes, and since the relevant variables are conserved, i.e., $\delta \mathbf{A}(\mathbf{0},t) = \delta \mathbf{A}(\mathbf{0})$ or equivalently $\delta \mathbf{\widetilde{A}}(\mathbf{0},\tau) = \delta \mathbf{\widetilde{A}}(\mathbf{0})$, the second term reduces to the standard Green-Kubo expression. In the realm of granular fluids the time dependence of $\langle \delta \mathbf{\widetilde{A}}(\mathbf{k},\tau) \delta \mathbf{\widetilde{A}}(-\mathbf{k}) \rangle_{H}^{-1}$ is consequential as it leads to non-negligible corrections [13].

Consider first the Navier-Stokes contributions to the energy dissipation rate. In the second term in Eq. (11), we need the entity:

$$\langle \delta \widetilde{p}_{\beta}(\mathbf{0},\tau) \, \delta \widetilde{A}^{a}_{\alpha}(\mathbf{0}) \rangle_{H} = \exp(\gamma_{H}\tau) \langle \delta \widetilde{p}_{\beta}(\mathbf{0}) \, \delta \widetilde{A}^{a}_{\alpha}(\mathbf{0}) \rangle_{H}$$
$$= \exp(\gamma_{H}\tau) V \frac{m^{2}n}{2} \, \delta_{\beta,\alpha} \delta_{a,p}, \quad (12)$$

where momentum conservation was used in the first equality. The factor $\exp(\gamma_H \tau)$ reflects the relative instability of the conserved quantity $\delta G^{\beta}(\mathbf{0})$ as compared to the decaying thermal velocity $v_0(t)$ (a fact that is related to the spontaneous appearance of vortices and clustering in undriven granular fluids [11,12]). To first order in ϵ , we then find

$$\langle \Gamma^{e}(\mathbf{r},t) \rangle_{NE} = \langle \Gamma^{e}(\mathbf{r},t) \rangle_{H} - \frac{2l_{0}v_{0}^{2}(t)}{mV} \int_{0}^{\infty} d\tau \exp(-\gamma_{H}\tau)$$
$$\times \langle \widetilde{\Gamma}_{D}^{e}(\mathbf{0},\tau) \widetilde{J}(\mathbf{0}) \rangle_{H} \nabla \cdot \mathbf{u}(\mathbf{r},t).$$
(13)

where $J(\mathbf{0}) \equiv \frac{1}{3} J_{\alpha\alpha}^{p}(\mathbf{0})$ (the trace of the momentum current). In the dilute limit the second term in Eq. (13), which is $\mathcal{O}(\epsilon) \nabla \cdot \mathbf{u}$, vanishes due to the fact that $J(\mathbf{0}) = (2/3)E(\mathbf{0})$ has no dissipative part. Density corrections due to collisional transfer give rise to a dissipative part of the pressure. These observations are consistent with results from Chapman-Enskog expansions of the Boltzmann [3,5] and the Enskog-Boltzmann equation [14]. Applying Eq. (11) to the energy current density $\langle \mathbf{J}^{e}(\mathbf{r},t) \rangle_{NE}$, we recover, e.g., the $\mathcal{O}(\epsilon)\nabla n$ contribution, whose existence is well-known from Chapman-Enskog expansion results [3,5]. Consider next the expression for the shear viscosity, for which Eq. (11) yields

$$\eta = \frac{2l_0 v_0(t)}{mV} \int_0^\infty d\tau \exp(-\gamma_H \tau) \langle \tilde{I}(\tau) \tilde{I} \rangle_H, \qquad (14)$$

where $\tilde{J}_{xyD}^{p}(\mathbf{0}) = \tilde{J}_{xy}^{p}(\mathbf{0}) \equiv \tilde{I}$. A (resummed) perturbative calculation of the shear viscosity [15,16] involves the splitting of \tilde{I} into a kinetic part, $\tilde{I}_{K} = m \Sigma_{i}^{N} c_{ix} c_{iy}$, and a collisional transfer part. Only the contribution of the kinetic part survives in the dilute limit, in which case the resulting value of η equals the Chapman-Enskog value. An efficient nonperturbative method for computing the Green-Kubo expression for finite densities is afforded by molecular dynamics simulations. This is relegated to a future publication.

An important issue not discussed above is the convergence problem induced by the existence of long-time tails. The latter can be especially harmful for the higher-ordergradient terms (though they are, most probably, rather small). This important issue will be taken up elsewhere.

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