Hydrodynamics of granular gases and granular gas mixtures

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It is shown that a vertically vibrated binary granular gas mixture of light and heavy particles can segregate (in the presence of gravity) in such a way that the bottom and top layers are composed mostly of light particles, even if all other parameters (including size) are the same for both species. The corresponding concentration profile possesses certain universal properties. It is also shown that such mixtures can segregate when the only difference between the species is the value of the coefficient of restitution. These findings follow from a set of hydrodynamic equations for granular gas mixtures which we derived from the pertinent Boltzmann equation. The above results comprise the second part of this article, the first part of which is devoted to a brief and somewhat biased review of the main physical properties of granular gases. This includes their (generic) tendency to coagulate into clusters and other micro- and macrostructures. A fundamental property of granular materials in general, and granular gases in particular, is the lack of scale separation; an explanation and some consequences are presented. The answer to the basic question of whether the dynamics of granular gases lends itself to description by (appropriate) hydrodynamic equations seems to be positive, though some restrictions apply.

1. Introduction

The fact that collections of macroscopic grains, such as sand, salt, cereals or coal, can flow is no surprise to anyone. Whether these flows can be described by hydro-dynamic equations is not *a priori* clear (Tan & Goldhirsch 1998; Kadanoff 1999; Goldhirsch 2000).

Collections of macroscopic grains, also known as granular matter, are commonplace in nature and industry. The handling, conveying and storage of grains (and the frequent malfunction of grain handling facilities, or even potentially catastrophic events such as the collapse of a silo) are of great industrial importance. Snow avalanches, rock and land slides, and sand dune dynamics, are examples of naturally occurring granular flows. The planetary rings are mostly composed of ice particles.

In addition to the practical and environmental importance of granular materials, there are excellent scientific reasons to study them. On one hand, fluidized granular materials exhibit almost every known hydrodynamic flow (and instability), such as Rayleigh–Bénard convection, Taylor–Couette flows, Faraday crispations, shear flows and more. On the other hand they possess a rich rheology which does not parallel

'regular' hydrodynamics. For instance, vertically vibrated shallow layers of grains exhibit 'oscillons' (Umbanhowar, Melo & Swinney 1996) which are stable geyser-like excitations. Granular fluids often posses non-trivial microstructures, which affect their other properties (Goldhirsch 2003). The normal stress in these fluids is often anisotropic, much like in other non-Newtonian materials (but for a somewhat different reason, see more below). Their unique properties led some to consider them to constitute 'a new state of matter'. To the theoretician granular materials pose significant challenges, many of which pertain to the fact that they lack scale separation (Tan & Goldhirsch 1998); in a way their hydrodynamic description can be considered to be an extreme case of the application of hydrodynamics (Goldhirsch 2000). Surprisingly, the field of granular flows became part of the discipline of fluid mechanics only about 25 years ago.

Often granular materials on the Earth are embedded in an ambient fluid. In this case the material is still considered to be granular if the stress due to the grains by far exceeds that due to the fluid (the ratio of the two is known as the Bagnold number), so that the effects of the fluid can be ignored; otherwise the system is to be considered a suspension.

Granular materials can be fluidized by means of vibration, shaking, shear, gravity (as in a chute) and other techniques (cf. the review Goldhirsch 2003). When strongly forced a granular system can become sufficiently fluidized that the grain interactions are mostly nearly instantaneous binary collisions; this phase is referred to as a granular gas.

Naturally occurring grains are usually non-spherical and polydisperse. While flowing or otherwise forced they are prone to attrition or breakup and their properties may therefore be time dependent. Furthermore, real grains experience frictional interactions (see e.g. Goldhirsch, Noskowicz & Bar-Lev 2005, and references therein). These complications notwithstanding, the theoreticians' paradigm for describing granular gases is one of a collection of smooth homogeneous hard spherical grains, whose collisions are characterized by a fixed coefficient of normal restitution. This model, in spite of its simplicity, brings out some of the major properties of granular gases (perhaps paralleling elastic spheres as models of molecular gases). Extensions include the use of (physically realistic) velocity-dependent coefficients of restitution, frictional restitution, and to a lesser extent non-sphericity (cf. the review Goldhirsch 2003). In the present article we employ the above simple model to describe the dynamics of monodisperse and bidisperse granular gases.

The structure of this paper is as follows. Section 2 is devoted to an introduction to the properties of granular gases. Section 3 provides an introduction to binary mixtures of granular gases. Section 4 is devoted to a description of the kinetics and hydrodynamics of these mixtures. Section 5 presents two novel results pertaining to segregation in binary granular gas mixtures. Finally, §6 provides a brief summary and outlook.

2. Some basic properties of granular gases

The theoretical description of granular gases is largely (but not entirely) based on analogies with molecular gases, where the grains are equivalents of the molecules.

There are two major differences between (classical) molecular and granular gases. As a practical matter, since typical grains are of macroscopic dimensions, the number of grains in an industrial container or experimental system is far smaller than Avogadro's number, rendering fluctuations of greater relative magnitude than in molecular gases.

There is an experimental advantage to the size of the grains since they are usually visible to the naked eye and fast photography can be employed (in particular in two-dimensional cases) to follow their dynamics. In such systems one can 'see' the interior of shocks (Rericha *et al.* 2002), whose widths may be e.g. of the order of centimetres. This fortunate state of affairs may cause (at times) some confusion, as hydrodynamic descriptions are often 'required' to describe the dynamics on the scale of a few grains; surprisingly, such applications seem to succeed in some cases, e.g. Forterre & Pouliquen (2002).

The fundamental difference between molecular and granular gases is that in the latter the collisions are inelastic. This fact alone is the source of many properties which are specific to granular gases (see below). The most obvious consequence of inelasticity is that granular gases 'do not conserve energy' (it is lost to internal degrees of freedom of the grains) in collisions, hence a steady state of a granular gas can only be sustained by an external energy supply. Therefore granular gases are always in non-equilibrium states.

At this stage it is convenient to define the granular temperature, T, as the (ensemble) average of the square of the fluctuating particle velocities. This temperature is unrelated to the internal (thermodynamic) temperature of the grains. It is however a measure of velocity fluctuations, much like the thermodynamic temperature for molecular gases. Notice that the dimension of the granular temperature is that of squared velocity.

Granular gases possess a tendency to coagulate into clusters even for 'initially prepared' homogeneous mass distributions (Goldhirsch 1991; Hopkins & Louge 1991; Goldhirsch & Zanetti 1993). This property can be understood on the basis of the following simple considerations, as well as detailed calculations (Goldhirsch & Zanetti 1993). A granular gas, like any other many-body system, experiences fluctuations, in particular of the density. Since in relatively dense domains the rate of collisions (proportional to the square of the number density) is higher than in dilute domains, the kinetic energy in dense domains decays at a higher rate than in dilute ones, owing to inelasticity. The ensuing (grain) pressure difference leads grains from dilute into dense domains, thereby further increasing the density of the latter, and giving rise to dense clusters. The clusters may further coagulate into larger clusters by coarsening (Goldhirsch 2003 and references therein) and they may collide thereby destroying each other (Tan & Goldhirsch 1997). Other structures or microstructures, such as plugs in sheared flows, can be attributed to similar mechanisms. Sufficiently small granular systems do not exhibit clustering, but even then there are linear instabilities that render them inhomogeneous (Goldhirsch & Zanetti 1993; Deltour & Barrat 1997; Brey, Ruiz-Monterro & Cubero 1999). Clustering is one of the reasons for multistability in granular gases, since they can be rather stable once formed.

Perhaps the most important property of granular gases, one which strongly distinguishes them from molecular gases, is the lack of scale separation (Tan & Goldhirsch 1998). Arguments that help demonstrate this property of granular gases in a simple case are presented next. Consider a simple shear flow of a monodisperse collection of spheres, with a fixed coefficient of normal restitution, $\tilde{e}$ (see equation (2.1)). The velocity field is given by $\boldsymbol{V} = \gamma y \hat{\boldsymbol{x}}$, where γ is the shear rate, x is the streamwise coordinate, and y a spanwise coordinate. In the absence of gravity, γ^{-1} provides just a time scale. Recalling that the granular temperature, T, has dimensions of squared velocity, it follows from dimensional analysis (also, from kinetic calculations or, by a direct comparison of the 'heating term' ($\propto \gamma^2 \ell \sqrt{T}$) in the equation of motion for the temperature field (see more below) to the energy sink term ($\propto (\varepsilon/\ell) T^{3/2}$)

in that equation) that $T \propto \gamma^2 \ell^2$, where ℓ is the mean free path (the only relevant microscopic length scale). Define the *degree of inelasticity*, ε, by $\varepsilon \equiv 1 - \tilde{e}^2$. Clearly, T should be larger, for a given value of γ, the smaller ε. Furthermore, when $\varepsilon = 0$, the shear work raises T indefinitely (the granular 'heat' cannot escape through the boundaries of the system). Therefore $T \to \infty$ as $\varepsilon \to 0$. It is plausible to assume (and this is corroborated by quantitative studies, cf. Sela & Goldhirsch (1998) and references therein; see also the above comparison of the heating and sink terms) that $T \propto 1/\varepsilon$, hence: $T = C(\gamma^2 \ell^2/\varepsilon)$. The value of C is about 1 in two dimensions and 3 in three dimensions. It follows that $\gamma \ell/\sqrt{T} = \sqrt{\varepsilon}/\sqrt{C}$, i.e. the change of speed over a mean free path (in the spanwise direction) is comparable to the thermal speed (unless ε is very small), hence the shear rate can be considered 'large', and the flow is typically supersonic. Indeed, shocks are a frequent occurrence in granular gases. Consider next the mean free time, τ, i.e. the ratio of the mean free path and the thermal speed: $\tau \equiv \ell/\sqrt{T}$. Clearly, in simple shear flows, τ and γ^{-1} are the microscopic and macroscopic time scales respectively, characterizing the system. Since $\tau/\gamma^{-1} = \tau \gamma = \ell \gamma/\sqrt{T} = \sqrt{\varepsilon}/\sqrt{C}$, there is no good temporal scale separation except in near-elastic cases. This fact may be used to cast some doubt on the relevance of granular hydrodynamic equations or methods to obtain them, which are based on the assumption of 'fast local equilibration' on microscopic scales, constrained by the values of the 'slow' or hydrodynamic fields. However, it does not preclude the existence of hydrodynamic descriptions of nearly elastic granular gases, where scale separation is restored. An analysis similar to the above for e.g. vertically vibrated granular systems (in the presence of gravity) reveals weak scale separation as well, but as it is less straightforward it will not be presented here.

An interesting consequence of the lack of scale separation is the anisotropy of the normal stress in granular gases. Consider again the simple shear flow. Since the gradients in this system are not 'small' one expects sizeable Burnett contributions to the constitutive relations. Consider the ratio of the xx component of the stress, τ_{xx}, and the pressure, p. As this is a (γ-dependent) dimensionless entity, which must be even in γ by symmetry, it follows that to $O(\gamma^2)$:

$$\frac{\tau_{xx}}{p} = \frac{1}{3}\left(1 + c_{xx}\frac{\gamma^2 \ell^2}{T}\right) = \frac{1}{3}\left(1 + \frac{c_{xx}\varepsilon}{C}\right),$$

in three dimensions. A similar result holds for τ_{yy}. The dimensionless constants, c_{xx} and c_{yy}, need not be equal; indeed they can be shown (Sela & Goldhirsch 1998) to be $O(1)$ and different from each other, i.e. one expects significant normal stress differences. Note that $\gamma^2 \ell^2/T \approx 10^{-18}$ for air at STP conditions and $\gamma = 0.1 \text{ s}^{-1}$. This shows yet another facet of granular gases: they may 'amplify' some negligible effects (in molecular gases) to the level of $O(1)$ phenomena.

2.1. *A brief review of the kinetic theory of granular gases*

It is easy to modify the classical Boltzmann equation to account for inelastic collisions (Goldshtein & Shapiro 1995). The assumption of molecular chaos, even for low densities, is not as well justified for granular as for molecular gases, since the normal component of the relative velocity is reduced by a factor $\tilde{e}$ upon collision, thereby rendering the particle velocities more correlated. Indeed, such correlations have been observed in molecular dynamic (MD) simulations, cf. e.g. Soto, Piasecki & Mareschal 2001. As the coefficient of restitution approaches unity, these correlations decrease. This implies that the Boltzmann equation for granular gases applies (at best) to near-elastic collisions. Since the Chapman–Enskog expansion is valid only when there

is scale separation, it follows that both granular kinetics and hydrodynamics are restricted, in principle, to near-elastic collisions.

Consider a monodisperse collection of homogeneous hard spheres, of unit mass, and diameter d, whose collisions are characterized by a constant coefficient of normal restitution, $\tilde{e}$. The binary collision between spheres i and j is defined by

$$v_i = v'_i - \frac{1+\tilde{e}}{2}(\hat{k} \cdot v'_{ij})\hat{k}, \qquad (2.1)$$

where (v'_i, v'_j) are the precollisional velocities, (v_i, v_j) are the corresponding postcollisional velocities, $v'_{ij} \equiv v'_i - v'_j$, and $\hat{k}$ is a unit vector pointing from the centre of sphere i to that of sphere j at the moment of contact. Denote by $f(v, r, t)$ the single-particle distribution function, i.e. the number density of particles having velocity v at point r and time t. The Boltzmann equation, corresponding to the above model, is (in three dimensions):

$$\frac{\partial f(v_1, r, t)}{\partial t} + v_1 \cdot \nabla f(v_1, r, t)$$
$$= d^2 \int_{\hat{k} \cdot v_{12} > 0} dv_2 \, d\hat{k} \, (\hat{k} \cdot v_{12}) \left(\frac{1}{\tilde{e}^2} f(v'_1, r, t) f(v'_2, r, t) - f(v_1, r, t) f(v_2, r, t) \right), \quad (2.2)$$

where ∇ is the gradient operator. In addition to the explicit dependence of (2.2) on $\tilde{e}$, it also implicitly depends on it through relation (2.1). The condition $\hat{k} \cdot v_{12} > 0$ represents the fact that only particles that approach each other can collide.

The Boltzmann equation is a nonlinear integrodifferential equation, and there is no known way to solve it exactly. Among the methods employed to obtain constitutive relations from the (granular) Boltzmann equation are phenomenological methods based on an ansatz for the form of the distribution function and the use of the Enskog equations for the moments of the distribution (cited in Goldhirsch 2003), the Grad expansion method and its generalizations (Jenkins & Richman 1985; Ramirez *et al.* 2000; Bisi, Spiga & Toscani 2004) and the Chapman–Enskog expansion (Chapman & Cowling 1970). The latter is the standard method used for the study of molecular gases, and it is based on an assumed existence of scale separation between the microscopic and macroscopic dynamics. The basic step in this expansion is to identify a set of 'slow' or hydrodynamic fields (usually the densities of the collisionally conserved entities) and assume that all other variables, in particular the velocity distribution function, are enslaved to the hydrodynamic fields. Technically, one replaces the space–time dependence of the distribution function, f, by a dependence on the (time-dependent) hydrodynamic fields. The next step is to assume that the fields do not change much over the scale of a mean free path (in space); this justifies a gradient expansion in the fields (formally, an expansion in powers of the Knudsen number, which is the ratio of the mean free path to the macroscopic scale of change of the fields). In practice, one can use a set of fields which are functionals of the slow fields, such as the velocity field. In elastic monodisperse systems the set of slow or hydrodynamic fields consists of the mass density, $\rho(r, t)$ (or number density, $n(r, t)$), the momentum density $p(r, t)$ (hence, one can use the velocity field, $V(r, t) \equiv p(r, t)/\rho(r, t)$ instead), and the energy density (for rigid objects, this is equivalent to the kinetic energy density; by subtracting the macroscopic kinetic energy density, $\frac{1}{2}\rho V^2$, from the 'full' energy density one obtains the fluctuating energy density, or $\frac{1}{2}\rho T$).

In the realm of granular gases, n and p are 'slow', but energy is not conserved (which is perhaps one of the reasons the granular temperature was not used as a

relevant field in early studies of granular dynamics, see Hutter & Rajagopal (1994) and references therein). However, when the degree of inelasticity is sufficiently small, it is justified to include it (or the granular temperature, $T(\boldsymbol{r}, t)$) as a hydrodynamic field, as it is 'nearly slow' (note that the granular temperature is eliminated from the equations of motion in some cases (Kumaran 2004; Wakou, Brito & Ernst 2002) as it is considered to relax to a slowly evolving 'steady' value when the spatial and temporal scales are sufficiently large). Furthermore, it is an important characterization of the state of a granular gas. Yet another argument for including the granular temperature in the set of the hydrodynamic fields is presented below. The aforementioned fields are moments of the distribution function, f:

$$n(\boldsymbol{r}, t) \equiv \int \mathrm{d}\boldsymbol{v}\, f(\boldsymbol{v}, \boldsymbol{r}, t), \quad \boldsymbol{V}(\boldsymbol{r}, t) \equiv \frac{1}{n(\boldsymbol{r}, t)} \int \mathrm{d}\boldsymbol{v}\, \boldsymbol{v}\, f(\boldsymbol{v}, \boldsymbol{r}, t),$$

and

$$T(\boldsymbol{r}, t) \equiv \frac{1}{n(\boldsymbol{r}, t)} \int \mathrm{d}\boldsymbol{v}(\boldsymbol{v} - \boldsymbol{V})^2 f(\boldsymbol{v}, \boldsymbol{r}, t),$$

respectively. It is *a priori* unclear whether these fields are sufficient for the achievement of a proper closure, since one cannot naively extrapolate from the case of molecular gases, but this turns out to be the case. The form of the (continuum mechanics) equations satisfied by the hydrodynamic fields can be obtained by multiplying the Boltzmann equation, (2.2), by 1, $\boldsymbol{v}_1$ and v_1^2 respectively, and integrating over $\boldsymbol{v}_1$. A standard procedure (Chapman & Cowling 1970), which exploits the symmetry properties of the collision integral on the right-hand side of the Boltzmann equation, yields (Sela & Goldhirsch 1998)

$$\frac{\mathrm{D}n}{\mathrm{D}t} + n\frac{\partial V_i}{\partial r_i} = 0, \tag{2.3}$$

$$n\frac{\mathrm{D}V_i}{\mathrm{D}t} + \frac{\partial P_{ij}}{\partial r_j} = 0, \tag{2.4}$$

$$n\frac{\mathrm{D}T}{\mathrm{D}t} + 2\frac{\partial V_i}{\partial r_j}P_{ij} + 2\frac{\partial Q_j}{\partial r_j} = -n\Gamma, \tag{2.5}$$

where $\mathrm{D}/\mathrm{D}t$ denotes the material derivative, and the summation convention is applied. Denote by $\boldsymbol{u} \equiv \boldsymbol{v} - \boldsymbol{V}$ the fluctuating (or peculiar) velocity. In (2.3)–(2.5) $P_{ij} \equiv n\langle u_i u_j \rangle$ denotes the stress tensor, and $Q_j \equiv \frac{1}{2}n\langle u^2 u_j \rangle$ is the heat flux vector, where $\langle\rangle$ denotes an average with respect to f. The term Γ, which accounts for the energy loss in the collisions, is given by:

$$\Gamma = \frac{\pi(1 - e^2)d^2}{8n} \int \mathrm{d}\boldsymbol{v}_1\, \mathrm{d}\boldsymbol{v}_2 v_{12}^3 f(\boldsymbol{v}_1) f(\boldsymbol{v}_2).$$

Equations (2.3)–(2.5) are exact consequences of the Boltzmann equation. They also comprise the equations of continuum mechanics, and thus their validity is more general than that of the Boltzmann equation. Notice that the only new term in the form of the continuum mechanics equations for granular fluids is the energy sink, Γ, in (2.5). This term is crucial for the correct description of granular hydrodynamics. Inelasticity also affects the detailed constitutive relations. When the latter are derived from the Boltzmann equation, their validity is limited to low volume fractions; the use of the Enskog–Boltzmann equation may yield constitutive relations which are valid up to moderate volume fractions, cf. e.g. Garzo & Dufty (1999a).

The zeroth order of the classical Chapman–Enskog perturbation theory (for elastically colliding constituents) corresponds to zero Knudsen number, K (no gradients), i.e. it is the state of (local) equilibrium, which is itself a solution of the Boltzmann equation. In the absence of an unforced, gradient-free granular gas steady state, a different perturbative scheme needs to be devised. One way (Brey *et al.* 1998) is to note the existence of a homogeneous solution of the Boltzmann equation, whose granular temperature decays with time (the 'homogeneous cooling state' or HCS (Goldhirsch, Noskowicz & Bar-Lev 2003)) and expand around the local HCS in powers of the Knudsen number. Another method is based on the observation that in the (double) elastic and unforced limit the equation possesses an equilibrium solution. Therefore, a double expansion in both the degree of inelasticity, ε, and K can be carried out. The former method does not formally restrict the value of ε to be small. The latter method is easier to implement, and it can be employed to produce any desired accuracy for the transport coefficients, though (in practice) only to low orders in ε. This restriction may not be too severe, since, as explained above, the Chapman–Enskog expansion cannot be valid for too small coefficients of restitution. For details on the constitutive relations obtained by using the second method the reader is referred to Sela & Goldhirsch (1998). Here we quote only the expression for the heat flux:

$$\boldsymbol{Q} = -\tilde{\kappa} n \ell \sqrt{T} \nabla T - \tilde{\lambda} \ell \sqrt{T^3} \nabla n + \text{Burnett terms},$$

where $\tilde{\kappa} \approx 0.4101 + 0.1072\varepsilon + O(\varepsilon^2)$ and $\tilde{\lambda} \approx 0.2110\varepsilon + O(\varepsilon^2)$. Notice that the heat flux includes a 'non-Fourier' term at order $K\varepsilon$. This term (as a low-density contribution) was first discovered in Sela & Goldhirsch (1998), rediscovered shortly thereafter in Brey *et al.* (1998), and further rediscovered on the basis of numerical simulations in Soto, Mareschal & Risso (1999). It is of importance in applications, e.g. in the determination of the temperature and density profiles of a vertically vibrated granular system (Brey, Ruiz-Montero & Moreno 2001).

Returning to the issue of choosing the slow fields for which hydrodynamic equations need to be derived, it should be evident on the basis of the above considerations that these fields are not necessarily the exactly conserved fields, but rather those fields that are conserved in the limit which defines the zeroth-order solution (this is compatible with the centre-manifold theorem, but we shall not dwell on this point here); in our case, when $\varepsilon \to 0$, the standard hydrodynamic fields become conserved.

3. Binary granular mixtures: a brief introduction

Granular mixtures are known to exhibit a host of effects that are specific to them, as well as other effects that pertain to mixtures in general. For instance, mass and size segregation are quite common, see e.g. the review Kudrolli (2004), the latter being the granular equivalent of the Soret effect, when driven by a temperature gradient. Typically, the tendency of granular mixtures to segregate (Ottino & Khakhar 2000; Shinbrot & Muzzio 2000; Kudrolli 2004) seems to be stronger than that of the corresponding molecular mixtures. For instance, granular mixtures may segregate on the basis of different frictional properties of the constituents (Kondic *et al.* 2003). Another interesting mechanism for segregation is ratcheting (Rapaport 2001). An intriguing phenomenon is the 'Brazil nut' effect, whereby a relatively large particle in a vibrated granular system tends to climb to the 'top' of the system, against gravity (Rosato *et al.* 1987). Several explanations of this phenomenon have been proposed, (cf. e.g. Rosato *et al.* 1987; Knight *et al.* 1996; Shinbrot & Muzzio 2000;

Trujillo, Alam & Herrmann 2003; Bose, Nott & Kumaran 2004; Mobius *et al.* 2005), some of which attribute the effect to the presence of convection (Knight *et al.* 1996).

While all of the proposed mechanisms may be relevant, it is interesting to note that the granular temperature of a vertically vibrated granular system decays as a function of height (distance from the source of energy, i.e. the floor) owing to the inelasticity of the particle interactions; this temperature gradient can induce a thermophoretic effect (Goldhirsch & Ronis 1983*a*, *b*), whereby a large particle moves down the temperature gradient. This possibility suggests that (at least) one of the mechanisms underlying the Brazil nut effect could be an indirect result of inelasticity in the sense that it is a thermophoretic phenomenon driven by the inelasticity-induced temperature gradient (convection in vertically vibrated granular systems is yet another example of an indirect consequence of inelasticity; it is also strongly affected by other factors, such as the nature of the sidewalls). Below we study a direct consequence of inelasticity, namely that a granular mixture, in which the constituents differ only by their respective coefficients of restitution, can segregate in the presence of a temperature gradient. Numerical and analytical studies of the dynamics of free ('cooling') granular mixtures, (e.g. Cattuto & Marconi 2004; Garzo & Dufty 1999*b*) suggest the existence of instabilities that may lead to segregation; however, (see below) a segregated state may be the basic solution of the equations describing a forced granular mixture. Many other interesting properties of granular mixtures, such as the patterns they exhibit (cf. e.g. Shinbrot & Muzzio 2000), possible segregation phase transitions (Reis & Mullin 2002), and segregation in sheared and other flows, are not described here.

The constituents of a granular mixture do not share the same (granular) temperature (Dahl *et al.* 2002; Feitosa & Menon 2002; Wildman & Parker 2002; Alam & Luding 2003; Paolotti *et al.* 2003; Wang, Jin & Ma 2003; Alam & Luding 2005). This much discussed property is not surprising since equipartition is not expected to hold in non-equilibrium states.

While models for mixtures of granular gases have been proposed, there are very few basic derivations of the pertinent constitutive relations. The first basic study of this problem seems to be Jenkins & Mancini (1989). These authors employed exact relations (Enskog equations) for the time evolution of single-particle properties. A closure was obtained by assuming a form of the velocity distribution function. A later study of granular thermal diffusion using a revised Enskog theory can be found in Hsiau & Hunt (1996). The resulting constitutive relations were essentially the same as for elastic systems, see Kincaid, Cohen & Lopez de Haro (1987) and references therein, except for an energy sink term in the equation for the granular temperature. Later, Arnarson & Willits (1998) improved upon these results, and used them to study segregation in a binary mixture of spheres subject to a temperature gradient. Further extensions are presented in Alam *et al.* (2002) and Jenkins & Yoon (2002). A smplified theory is proposed in Arnarson & Jenkins (2004). A critical study of the virtues and disadvantages of a series of models for the dynamics of granular binary mixtures alongside a comparison to results of molecular dynamic simulations of shear flows (cf. e.g. Alam & Luding 2005; Clelland & Hrenya 2002) can be found in (Galvin, Dahl & Hrenya 2004). In a relatively recent study (Garzo & Dufty 2002) the Boltzmann equation for granular mixtures was perturbatively solved to obtain constitutive relations, using the local HCS as a zeroth order. As the algebra involved in these derivations is complex, the constitutive relations are not explicitly presented in their paper. Our own work, which is nominally limited to near-elastic granular gases, involves some complex algebra as well, but by using a computer-aided technique we

have been able to obtain accurate analytic expressions for the constitutive relations, to linear order in the degress of inelasticity. In the near-elastic case our results agree with those of Garzo & Dufty.

The outline of the derivation of constitutive relations for granular mixtures presented below is based on the above generalization of the Chapman–Enskog expansion, which involves an expansion around local equilibrium. Therefore, the hydrodynamic fields are the densities of the entities that are conserved in the zeroth order of perturbation theory, i.e. the same as in an elastically interacting mixture.

4. Kinetics and hydrodynamics of granular gas mixtures

Consider a mixture of smooth hard spheres, composed of species A and B, of masses m_A and m_B, and diameters σ_A and σ_B, respectively. The coefficient of normal restitution (assumed to be fixed) for a collision of a particle of species $\alpha \in \{A, B\}$ with a particle of species $\beta \in \{A, B\}$ is denoted by $e_{\alpha\beta}$ (hence, $e_{\alpha\beta} \in \{e_{AA}, e_{BB}, e_{AB}\}$). Tangential (frictional) restitution is not considered here. The transformation of velocities due to a collision of a sphere of species α with a sphere of species β is given by

$$\boldsymbol{v}_1 = \boldsymbol{v}'_1 - (1 + e_{\alpha\beta})M^{\beta,\alpha}(\boldsymbol{v}'_{12} \cdot \hat{\boldsymbol{k}})\hat{\boldsymbol{k}}, \tag{4.1}$$

$$\boldsymbol{v}_2 = \boldsymbol{v}'_2 + (1 + e_{\alpha\beta})M^{\alpha,\beta}(\boldsymbol{v}'_{12} \cdot \hat{\boldsymbol{k}})\hat{\boldsymbol{k}}, \tag{4.2}$$

where $\{\boldsymbol{v}'_1, \boldsymbol{v}'_2\}$ denote the precollisional velocities of the spheres (the index '1' refers here to species α), and $\{\boldsymbol{v}_1, \boldsymbol{v}_2\}$ are the corresponding postcollisional velocities; $\hat{\boldsymbol{k}}$ is a unit vector pointing from the centre of sphere α to that of sphere β, $M^{\alpha,\beta} \equiv m_\alpha/(m_\alpha + m_\beta)$ and $\boldsymbol{v}_{12} \equiv \boldsymbol{v}_1 - \boldsymbol{v}_2$, a similar definition holding for the primed (precollisional) velocities. Obvious kinematic constraints require that $\boldsymbol{v}'_{12} \cdot \hat{\boldsymbol{k}} \geqslant 0$. Define the degrees of inelasticity, corresponding to the coefficients of restitution, as $\varepsilon_{\alpha\beta} \equiv 1 - e_{\alpha\beta}^2$.

The kinetic description of a binary granular gas mixture involves two Boltzmann equations, one for each species. Their derivation is similar to that of (2.2), and will not be presented here; see also Garzo & Dufty (2002). The result is

$$\frac{\partial f_\alpha}{\partial t} + \boldsymbol{v}_1 \cdot \nabla f_\alpha + \boldsymbol{g} \cdot \nabla_{v_1} f_\alpha = \mathscr{B}_{\alpha\alpha}(f_\alpha, f_\alpha, e_{\alpha\alpha}) + \mathscr{B}_{\alpha\beta\ (\beta \neq \alpha)}(f_\alpha, f_\beta, e_{\alpha\beta}), \tag{4.3}$$

where $\boldsymbol{g}$ denotes the gravitational acceleration, ∇_{v_1} is a gradient with respect to the vector $\boldsymbol{v}_1$, $f_\alpha(\boldsymbol{v})$ is the single-particle (or singlet) distribution function for particles of species α, and $\mathscr{B}_{\alpha\beta}(f_\alpha, f_\beta, e_{\alpha\beta})$ is the α-β species Boltzmann collision operator, defined by:

$$\mathscr{B}_{\alpha\beta}(f_\alpha, f_\beta, e_{\alpha\beta}) = \sigma_{\alpha\beta}^2 \iint_{v_{12} \cdot k > 0} \left[\frac{f_\alpha(\boldsymbol{v}'_1)f_\beta(\boldsymbol{v}'_2)}{e_{\alpha\beta}^2} - f_\alpha(\boldsymbol{v}_1)f_\beta(\boldsymbol{v}_2) \right] (\boldsymbol{v}_{12} \cdot \hat{\boldsymbol{k}}) \, \mathrm{d}\boldsymbol{v}_2 \, \mathrm{d}\hat{\boldsymbol{k}}, \tag{4.4}$$

where $\sigma_{\alpha\beta} \equiv (\sigma_\alpha + \sigma_\beta)/2$. Here $\boldsymbol{v}_1$ and $\boldsymbol{v}'_1$ pertain to species α, and $\boldsymbol{v}_2$ and $\boldsymbol{v}'_2$ pertain to β. Notice that $\mathscr{B}_{\alpha\beta}(f_\alpha, f_\beta, e_{\alpha\beta})$ depends on the coefficients of normal restitution both explicitly, as shown in (4.4), and implicitly through the collision law.

Following §3 the hydrodynamic fields in the present case comprise the two number densities n_A and n_B (or the mass densities $\rho_A = n_A m_A$ and $\rho_B = n_B m_B$), the mixture's velocity field, $\boldsymbol{V}$, and the temperature field, T, defined (differently from the mono-disperse case) as twice the mean fluctuating kinetic energy of a fluid particle. The continuum equations of motion follow from the pertinent Boltzmann equation directly

(using the standard procedure of computing velocity moments of the equation). Their validity is general since they are based on the underlying conservation laws.

The equation of motion for the number density, n_α, with $\alpha \in \{A, B\}$, is

$$\frac{\mathrm{D}n_\alpha}{\mathrm{D}t} = -\nabla \cdot \boldsymbol{J}_\alpha - n_\alpha \nabla \cdot \boldsymbol{V} \qquad (4.5)$$

where $\mathrm{D}/\mathrm{D}t$ is the material derivative and $\boldsymbol{J}_\alpha \equiv n_\alpha(\boldsymbol{V}_\alpha - \boldsymbol{V})$ is the particle flux density of species α. As $\boldsymbol{V}_\alpha$, the velocity field of species α (or the flux $\boldsymbol{J}_\alpha$), is not a hydrodynamic field, it must be given by an appropriate constitutive relation. The velocity field obeys (as expected)

$$\rho \frac{\mathrm{D}V_i}{\mathrm{D}t} = -\frac{\partial P_{ij}}{\partial x_j} + \rho \boldsymbol{g},$$

where the summation convention is used, and P_{ij} is the stress tensor. The granular temperature field obeys

$$n\frac{\mathrm{D}T}{\mathrm{D}t} = T\nabla \cdot \boldsymbol{J} - \nabla \cdot \boldsymbol{Q} - 2P_{ij}\frac{\partial V_i}{\partial x_j} - \Gamma,$$

where $\boldsymbol{J} \equiv \boldsymbol{J}_A + \boldsymbol{J}_B$ is the total particle flux, and $\boldsymbol{Q}$ is the heat flux.

The form of the constitutive relations can be easily determined from tensorial (and symmetry) considerations, the result being (to linear order in the gradients, or Navier–Stokes order)

$$P_{ij} = p\delta_{ij} - 2\mu D_{ij} - \delta_{ij}\eta\nabla \cdot \boldsymbol{V}, \quad \text{where } D_{ij} \equiv \frac{1}{2}\left(\frac{\partial V_i}{\partial x_j} + \frac{\partial V_j}{\partial x_i} - \frac{2}{3}\delta_{ij}\nabla \cdot \boldsymbol{V}\right)$$

is the traceless rate of strain tensor, μ is the shear viscosity, η is the bulk viscosity (which vanishes in the dilute limit) and p is the pressure. The diffusion flux is given by

$$\boldsymbol{J}_\alpha = -\frac{m_{\beta(\beta \neq \alpha)}n}{\rho}D\nabla\left(\frac{n_\alpha}{n}\right) - \frac{\rho}{p}D_p\nabla p - \frac{\rho}{p}D_T\nabla T, \qquad (4.6)$$

where D and D_T and D_p are the diffusion, thermal diffusion and barodiffusion coefficients respectively, and $\rho \equiv \rho_A + \rho_B$ is the mass density. The heat flux is given by

$$\boldsymbol{Q} = -T^2 D_f \nabla\left(\frac{n_A}{n}\right) - \lambda_p \nabla p - \lambda \nabla T, \qquad (4.7)$$

where D_f is the Dufour coefficient, λ is the thermal conductivity and λ_p is the barothermal coefficient. The energy sink term will be discussed further below. In the dilute limit the equation of state is the same as that for an ideal gas: $p = \frac{1}{3}nT$. For various other forms of the constitutive relations for $\boldsymbol{J}_\alpha$ and $\boldsymbol{Q}$ see e.g. Landau & Lifshitz (1959) and Garzo & Dufty (2002) and below; they are all trivially related to each other.

4.1. *Kinetic theoretical description*

Let $f_\alpha(\boldsymbol{v})$ denote the single-particle distribution function (the spatial and temporal dependence of f are suppressed) of species α. Following the standard definitions of kinetic theory, the number density for species α is given by $n_\alpha = \int f_\alpha(\boldsymbol{v})\,\mathrm{d}\boldsymbol{v}$, the corresponding mass density being $\rho_\alpha = m_\alpha n_\alpha$; the overall number density is $n \equiv n_A + n_B$, and the overall mass density is $\rho \equiv \rho_A + \rho_B$. The velocity field of species α is given by $\boldsymbol{V}_\alpha = (1/n_\alpha)\int f_\alpha(\boldsymbol{v})\boldsymbol{v}\,\mathrm{d}\boldsymbol{v}$. Note that $\boldsymbol{V}_\alpha$ is not a hydrodynamic field, and needs

to be expressed as a functional of the hydrodynamic fields. The mixture's velocity field is $V = (1/\rho)(\rho_A V_A + \rho_B V_B)$. The granular temperature of species α is defined by $T_\alpha = (1/n_\alpha) \int f_\alpha(v) m_\alpha (v - V)^2 \, dv$. Note that T_α is not a hydrodynamic field. The velocity fluctuations of each species are measured with respect to the (hydrodynamic) mixture's velocity field, not the species' velocity field. The mixture's granular temperature is defined as $T = (1/n)(n_A T_A + n_B T_B)$. The kinetic expression for the stress tensor is obtained by standard means, cf. e.g. Chapman & Cowling (1970), the result being

$$P_{ij} = m_A \int f_A(v) u_i u_j \, dv + m_B \int f_B(v) u_i u_j \, dv, \tag{4.8}$$

where $u \equiv v - V$ is the peculiar (fluctuating) velocity of a particle (irrespective of its species). Similarly, the heat flux is composed of two contributions, for obvious reasons:

$$Q = \int f_A(v) m_A u^2 u \, dv + \int f_B(v) m_B u^2 u \, dv. \tag{4.9}$$

Our definition of the heat flux may differ by a factor of 2 from some other definitions. The sink term is given by $\Gamma = \Gamma_A + \Gamma_B + \Gamma_{AB}$, where

$$\Gamma_\alpha \equiv \varepsilon_{\alpha\alpha} \frac{m_\alpha \pi \sigma_\alpha^2}{8} \iint f_\alpha(v_1) f_\alpha(v_2) |v_{12}|^3 \, dv_1 \, dv_2, \tag{4.10}$$

and

$$\Gamma_{AB} \equiv \varepsilon_{AB} \frac{m_{AB} \pi \sigma_{AB}^2}{2} \iint f_A(v_1) f_B(v_2) |v_{12}|^3 \, dv_1 \, dv_2. \tag{4.11}$$

4.2. *Outline of the Chapman–Enskog expansion*

We solved (4.3) perturbatively by using a generalized Chapman–Enskog expansion, following Sela & Goldhirsch (1998). The small parameters in this expansion are $\{\varepsilon_{\alpha\beta}\}$, and the Knudsen number, K. As mentioned, when $K = \varepsilon_{\alpha\beta} = 0$, the system comprises a free elastically colliding mixture, for which the Boltzmann equation admits an equilibrium solution. As is customary, the distribution functions of the mixture are written as

$$f_\alpha = f_\alpha^{(0)}(1 + \phi_\alpha), \quad \text{where } f_\alpha^{(0)} = n_\alpha \left(\frac{3m_\alpha}{2\pi T} \right)^{3/2} \exp \left(-\frac{3m_\alpha u^2}{2T} \right)$$

are local equilibrium distribution functions, and the corrections ϕ_α are expanded as follows: $\phi_\alpha = \phi_\alpha^K + \phi_\alpha^\varepsilon + \phi_\alpha^{K\varepsilon} + \phi_\alpha^{KK} + \phi_\alpha^{\varepsilon\varepsilon} + \dots$, where superscripts denote the order of each term in the small parameters (a superscript ε denoting all terms that are of the mentioned order in the three degrees of inelasticity). Following the Chapman–Enskog method, it is assumed that f_α is a functional of the hydrodynamic fields and that its time dependence is implicit, i.e. it is determined by the time dependence of these fields alone. Therefore, the time derivative of the distribution function can be expressed in terms of the time derivatives of the hydrodynamic fields:

$$\mathscr{D} f_\alpha = (1 + \phi_\alpha) f_\alpha^{(0)} \left[\mathscr{D} \ln n_\alpha + \frac{3m_\alpha u_i}{T} \mathscr{D} V_i + \left(\frac{3m_\alpha u^2}{2T} - \frac{3}{2} \right) \mathscr{D} \ln T \right] + f_\alpha^{(0)} \mathscr{D} \phi_\alpha, \tag{4.12}$$

where

$$\mathscr{D} f \equiv \frac{\partial f}{\partial t} + v \cdot \nabla f = \frac{Df}{Dt} + u \cdot \nabla f.$$

Recall that u is the fluctuating velocity of the particle, v its actual velocity, V is the velocity field and $D/Dt \equiv \partial/\partial t + V \cdot \nabla$ is the material derivative (notice that $\mathscr{D}$ is not

a material derivative). In parallel to the expansion of f, the result of application of the operator $\mathscr{D}$ to any functional of the fields and the (fluctuating) velocities, can be expanded in powers of the small parameters. Therefore one can define the operators $\mathscr{D}^0$, $\mathscr{D}^K$, $\mathscr{D}^{K\varepsilon}$, and so on, by the formal expansion $\mathscr{D}F = \mathscr{D}^0 F + \mathscr{D}^K F + \mathscr{D}^{K\varepsilon} F + \dots$ (for any function, F, of space, time and the phase space variables) where the superscripts identify the orders in the small parameters (see also Sela & Goldhirsch 1998). Next, upon expanding the Boltzmann equation (using the expansion of f_α and the Chapman–Enskog ansatz) in the small parameters and equating terms of the same order on both sides of the equation, one obtains a set of equations corresponding to each order in the perturbative expansion. For instance, the resulting equations at the zeroth and linear orders in ε are given by (with $\beta \neq \alpha$)

$$\mathscr{D}^0 f_\alpha = \mathscr{B}_{\alpha\alpha}\big(f_\alpha^{(0)}, f_\alpha^{(0)}, 1\big) + \mathscr{B}_{\alpha\beta}\big(f_\alpha^{(0)}, f_\beta^{(0)}, 1\big) = 0,$$

$$\mathscr{D}^\varepsilon f_\alpha = \mathscr{B}_{\alpha\alpha}\big(f_\alpha^{(0)}, f_\alpha^{(0)}\phi_\alpha^\varepsilon, 1\big) + \mathscr{B}_{\alpha\alpha}\big(f_\alpha^{(0)}\phi_\alpha^\varepsilon, f_\alpha^{(0)}, 1\big) + \mathscr{B}_{\alpha\beta}\big(f_\alpha^{(0)}, f_\beta^{(0)}\phi_\beta^\varepsilon, 1\big)$$

$$+ \mathscr{B}_{\alpha\beta}\big(f_\alpha^{(0)}\phi_\alpha^\varepsilon, f_\beta^{(0)}, 1\big) + \varepsilon_{\alpha\alpha}\left.\frac{\partial \mathscr{B}_{\alpha\alpha}\big(f_\alpha^{(0)}, f_\alpha^{(0)}, e_{\alpha\alpha}\big)}{\partial \varepsilon_{\alpha\alpha}}\right|_{e_{\alpha\alpha}=1}$$

$$+ \varepsilon_{\alpha\beta}\left.\frac{\partial \mathscr{B}_{\alpha\beta}\big(f_\alpha^{(0)}, f_\beta^{(0)}, e_{\alpha\beta}\big)}{\partial \varepsilon_{\alpha\beta}}\right|_{e_{\alpha\beta}=1}.$$

The action of the $\mathscr{D}$ operators on $f^{(0)}$ is found by using (4.12), and the expansion of the action of $\mathscr{D}$ on the hydrodynamic fields in powers of the small parameters is carried out using the continuum equations of motion presented above.

The form of the corrections ϕ_α^X, where $X \in \{\varepsilon_{\alpha\beta}, K, K\varepsilon_{\alpha\beta}, KK, \varepsilon_{\alpha\beta}\varepsilon_{\gamma\delta}, \dots\}$, is dictated by symmetry and tensorial considerations, though it also directly results from the obtained equations. For instance:

$$\phi_\alpha^K = \phi_\alpha^{K,T} \boldsymbol{u} \cdot \nabla \ln T + \phi_\alpha^{K,n_A} \boldsymbol{u} \cdot \nabla \ln n_A + \phi_\alpha^{K,n_B} \boldsymbol{u} \cdot \nabla \ln n_B + \phi_\alpha^{K,V} \overline{u_i u_j} \frac{\partial V_i}{\partial x_j},$$

to linear order in the gradients, where for any tensor $\overline{A}_{ij} \equiv \frac{1}{2}(A_{ij} + A_{ji} - \frac{2}{3}\delta_{ij}A_{kk})$, and the prefactors are scalars in $\boldsymbol{u}$; also $\phi_\alpha^{\varepsilon_{\alpha\beta}}$ is itself a scalar in $\boldsymbol{u}$. It is convenient to define the following entities:

$$\tilde{\phi}_\alpha^{\varepsilon_{\alpha\beta}} \equiv \phi_\alpha^{\varepsilon_{\alpha\beta}}, \quad \tilde{\phi}_{\alpha i}^{K,T} \equiv \phi_\alpha^{K,T} u_i, \quad \tilde{\phi}_{\alpha i}^{K,n_A} \equiv \phi_\alpha^{K,n_A} u_i, \quad \tilde{\phi}_{\alpha i}^{K,n_B} \equiv \phi_\alpha^{K,n_B} u_i, \quad \tilde{\phi}_{\alpha,i,j}^{K,V} \equiv \phi^{K,V} \overline{u_i u_j}$$

and so on, as the 'prefactors' of the gradients of the fields (which have explicit tensorial dependence). These entities, collectively denoted by $\tilde{\phi}_\alpha^X$, satisfy the following coupled equations (with $\alpha \in \{A, B\}$, $\alpha \neq \beta$):

$$\mathscr{L}_\alpha\big(\tilde{\phi}_\alpha^X(\boldsymbol{v}_1)\big) + \mathscr{L}_{\alpha\beta}\big(\tilde{\phi}_\alpha^X(\boldsymbol{v}_1) + \tilde{\phi}_\beta^X(\boldsymbol{v}_2)\big) = \mathscr{R}_\alpha^X\big(\boldsymbol{v}_1^2\big)\boldsymbol{H}_\alpha^X(\boldsymbol{v}_1), \tag{4.13}$$

where on the right-hand side $\mathscr{R}_\alpha^X\big(\boldsymbol{v}_1^2\big)$ is a scalar and $\boldsymbol{H}_\alpha^X(\boldsymbol{v}_1)$ is a tensor in $\boldsymbol{v}_1$ (determined by the equations of motion and the previous orders in the expansion), and where for any function of the velocity, ψ, or function of two velocity arguments χ, the linearized Boltzmann operators are defined by

$$\mathscr{L}_\alpha(\psi(\boldsymbol{v}_1)) \equiv \sigma_\alpha^2 \iint_{v_{12}\cdot k>0} f_\alpha^{(0)}(\boldsymbol{v}_1) f_\alpha^{(0)}(\boldsymbol{v}_2)(\boldsymbol{v}_{12}\cdot \hat{\boldsymbol{k}})$$

$$\times (\psi(\boldsymbol{v}_1') + \psi(\boldsymbol{v}_2') - \psi(\boldsymbol{v}_1) - \psi(\boldsymbol{v}_2))\,\mathrm{d}\boldsymbol{v}_2\,\mathrm{d}\hat{\boldsymbol{k}}, \tag{4.14}$$

$$\mathscr{L}_{\alpha\beta}(\chi(\boldsymbol{v}_1, \boldsymbol{v}_2)) \equiv \sigma_{\alpha\beta}^2 \iint_{v_{12}\cdot k>0} f_\alpha^{(0)}(\boldsymbol{v}_1) f_\beta^{(0)}(\boldsymbol{v}_2)(\boldsymbol{v}_{12}\cdot \hat{\boldsymbol{k}})$$

$$\times (\chi(\boldsymbol{v}_1', \boldsymbol{v}_2') - \chi(\boldsymbol{v}_1, \boldsymbol{v}_2))\,\mathrm{d}\boldsymbol{v}_2\,\mathrm{d}\hat{\boldsymbol{k}}. \tag{4.15}$$

It should be understood that the linearized Boltzmann operators are truly functionals of the functions they act upon (they are written above as if they were mere functions; e.g. strictly speaking $\mathscr{L}(\psi(\boldsymbol{v}_1))$ should read $\mathscr{L}(\{\psi(\boldsymbol{v})\};\boldsymbol{v}_1)$, i.e. the action of $\mathscr{L}$ on ψ evaluated at $\boldsymbol{v}_1$). Notice that the velocity transformations in the definition of $\mathscr{L}_{\alpha\beta}$ correspond to elastic collisions between the α and β spheres, with $\boldsymbol{v}_1$ pertaining to α. Since the linearized Boltzmann operator has vanishing eigenvalues, corresponding to the collisional invariants, 1 or m_α, $m_\alpha \boldsymbol{v}$ and $m_\alpha v^2$ (recall that the linearized Boltzmann operator is the same as in the elastic case; see also Chapman & Cowling (1970)), the general solution to these equations comprise a sum of particular solution and a linear combination of the summational invariants: $\phi_\alpha = \Phi_\alpha + a_{1,\alpha} + \boldsymbol{a}_{2,\alpha} \cdot (m_\alpha \boldsymbol{v}) + a_{3,\alpha}\, m_\alpha v^2$, where Φ_α is a particular solution, and the 'a' prefactors are independent of $\boldsymbol{v}$. Uniqueness is assured by requiring that the solutions obey the following orthogonality (which are also solubility) conditions to every order (X) in the perturbation expansion:

$$\int f_A \phi_A^X \, d\boldsymbol{v} = 0, \quad \int f_B \phi_B^X \, \mathrm{d}\boldsymbol{v} = 0, \quad \int f_A \phi_A^X (m_A \boldsymbol{v}) \, \mathrm{d}\boldsymbol{v} + \int f_B \phi_B^X (m_B \boldsymbol{v}) \, \mathrm{d}\boldsymbol{v} = 0,$$

$$\int f_A \phi_A^X (m_A v^2) \, \mathrm{d}\boldsymbol{v} + \int f_B \phi_B^X (m_B v^2) \, \mathrm{d}\boldsymbol{v} = 0.$$

These conditions ensure that the temperature, densities and hydrodynamic velocity that appear in the zeroth-order (local equilibrium) solution are consistent with the exact fields (as is standard in the Chapman–Enskog expansion), i.e. their values are not corrected by the perturbations, ϕ_α^X. In practice, these conditions apply to the scalar and vector corrections (in the peculiar velocity; the latter appear in scalar products with the gradients of the fields) only, and are translated to conditions on the (zeroth and first) coefficients that appear in the Sonine polynomial expansion presented below. The conditions are automatically satisfied for higher-order tensors, by symmetry. The next step is to expand the functions $\tilde{\phi}_\alpha^X$ in a (truncated) series of Sonine polynomials:

$$\tilde{\phi}_\alpha^X(u) = \sum_{n=0} \psi_\alpha^{X,(n)} S_m^n \left(\frac{3 m_\alpha u^2}{2T} \right) \boldsymbol{H}_\alpha^X(\boldsymbol{v}_1),$$

where $\psi_\alpha^{X,(n)}$ are the expansion coefficients. The index m depends on the tensorial rank of $\boldsymbol{H}_\alpha^X(\boldsymbol{v}_1)$: it equals $\frac{1}{2}$ for a scalar, $\frac{3}{2}$ for a vector, $\frac{5}{2}$ for a second-order tensor, and so on. This expansion is possible because the linear Boltzmann operator is isotropic, i.e. it preserves the tensorial nature and ranks of the tensors of the velocity it acts upon (it also produces traceless tensors when acting on traceless tensors; in this way different tensors of the same rank can be separated). Upon substituting the Sonine polynomial expansion in (4.13), and projecting both sides on each of the Sonine polynomials, one obtains a linear system of equations for the coefficients $\psi_\alpha^{X,(n)}$. Notice that the solutions depend on the order of truncation of the expansions. Owing to the heavy algebra involved in the calculations, only the lowest non-zero orders in the Sonine polynomial expansions have been considered in previous studies. By exploiting the fact that the Sonine polynomials are derivatives of corresponding generating functions, we were able to harness symbolic manipulators (such as Maple) to carry out the Chapman–Enskog expansion to practically any desired order of truncation. The solutions of the perturbative expansions, once obtained, serve to calculate the constitutive relations, using the above expressions for the fluxes and sink term. Notice that as the perturbative expansion is not affected by the gravitational term in (4.3) (see Chapman & Cowling (1970)), g does not enter the constitutive relations.

4.3. *Constitutive relations*

The dependence of the transport coefficients on the parameters of the problem is rather complex. Here we present an expression for the energy sink term and the form of the other constitutive relations (which can be guessed on the basis of tensorial considerations). The explicit dependence of the transport coefficients on the parameters is presented in the Appendix.† The diffusion fluxes, to Navier–Stokes order, are given by

$$\boldsymbol{J}_\alpha = \frac{1}{\sqrt{6}} \frac{n_\alpha}{n} \frac{1}{\sigma_{AB}^2} \sqrt{\frac{T}{m_\alpha}} \left(\kappa_\alpha^T \boldsymbol{\nabla} \ln T + \kappa_\alpha^{n_A} \boldsymbol{\nabla} \ln n_A + \kappa_\alpha^{n_B} \boldsymbol{\nabla} \ln n_B \right), \quad (4.16)$$

where κ_α^T, $\kappa_\alpha^{n_A}$ and $\kappa_\alpha^{n_B}$ are non-trivial functions of the set of dimensionless parameters

$$\mathscr{S} \equiv \left\{ \{\varepsilon_{\alpha\beta}\}, M_A \equiv \frac{m_A}{m_A + m_B}, \frac{\sigma_A}{\sigma_A + \sigma_B}, c \right\},$$

where the concentration field, c, is defined as $c \equiv n_A/n = n_A/(n_A + n_B)$. Explicit expressions for these coefficients are provided in the Appendix. It is trivial and convenient to rewrite (4.16) as follows:

$$\boldsymbol{J}_\alpha = \frac{1}{\sqrt{6}} \frac{n_\alpha}{n} \frac{1}{\sigma_{AB}^2} \sqrt{\frac{T}{m_\alpha}} \left(\kappa_\alpha^T \boldsymbol{\nabla} \ln T + \kappa_\alpha^n \boldsymbol{\nabla} \ln n + \kappa_\alpha^c \boldsymbol{\nabla} \ln c \right), \quad (4.17)$$

where one should recall that $\sigma_{AB} \equiv (\sigma_A + \sigma_B)/2$. The prefactors κ_α^T, κ_α^n and κ_α^c are trivially related to κ_α^T, $\kappa_\alpha^{n_A}$ and $\kappa_\alpha^{n_B}$. It is also trivial to transform either (4.16) or (4.17) to the often used form, (4.6). The heat flux is given by:

$$\boldsymbol{Q} = \frac{5\sqrt{6}}{18} \frac{1}{\sigma_{AB}^2} \frac{T^{3/2}}{\sqrt{m_0}} \left(\lambda^T \boldsymbol{\nabla} \ln T + \lambda^{n_A} \boldsymbol{\nabla} \ln n_A + \lambda^{n_B} \boldsymbol{\nabla} \ln n_B \right), \quad (4.18)$$

where $m_0 \equiv m_A + m_B$. Explicit expressions for the coefficients λ^T, λ^{n_A} and λ^{n_B} are presented in the Appendix. It is easy to transform (4.18) to the following form:

$$\boldsymbol{Q} = \frac{5\sqrt{6}}{18} \frac{1}{\sigma_{AB}^2} \frac{T^{3/2}}{\sqrt{m_0}} \left(\lambda^T \boldsymbol{\nabla} \ln T + \lambda^n \boldsymbol{\nabla} \ln n + \lambda^c \boldsymbol{\nabla} \ln c \right) \quad (4.19)$$

and relate λ^T, λ^n and λ^c to the prefactors that appear in (4.18). The sink term, to first order in the degrees of inelasticity, is given by

$$\Gamma = \frac{4\sqrt{3\pi}}{9} \left(\varepsilon_{AA} \frac{n_A^2 \sigma_A^2}{\sqrt{m_A}} + \varepsilon_{BB} \frac{n_B^2 \sigma_B^2}{\sqrt{m_B}} + \varepsilon_{AB} \sqrt{2} \frac{n_A n_B \sigma_{AB}^2}{\sqrt{m_{AB}}} \right) T^{3/2} \quad (4.20)$$

where $m_{AB} \equiv (m_A m_B)/(m_A + m_B)$. In the elastic limit the above constitutive relations reduce to well-known results, cf. e.g. Chapman & Cowling (1970), minor differences being due to the fact that we have carried out the expansion in the Sonine polynomials to third order (the result, to second order, can be found in the Appendix), whereas in previous studies only the first-order contribution was calculated. Similarly, our results agree with those of Garzo & Dufty (2002) in the common domain of validity (small $\varepsilon_{\alpha\beta}$).

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† The appendix is available as a supplement to the online version of this paper or on request from the authors.

5. Applications

5.1. *Inelasticity-driven segregation*

This subsection is devoted to the description of a novel effect, namely the fact that even when the species differ only by their respective coefficients of restitution, they segregate when subject to a temperature gradient. Consider a non-convecting ($V = 0$) steady state of a mixture for simplicity. Clearly, the diffusion flux vanishes in this state.

In this case, the equation for the momentum density reduces to $\nabla p = -\rho g$, which can be rewritten, using the equation of state, $p = \frac{1}{3}nT$, and $\rho = (m_A c + m_B(1-c))n$, as

$$\nabla \ln n = -\nabla \ln T - \frac{3}{T}((m_A - m_B)c + m_B)g. \tag{5.1}$$

Upon substituting (5.1) in (4.17), and using the fact that in a non-convecting state the diffusive fluxes vanish, i.e. $J_A = 0$, one obtains the following relation between the temperature and concentration gradients:

$$\nabla \ln c = \frac{\kappa_A^n - \kappa_A^T}{\kappa_A^c} \nabla \ln T + \frac{3}{T}\frac{\kappa_A^n}{\kappa_A^c}((m_A - m_B)c + m_B))g. \tag{5.2}$$

Define the rescaled length:

$$\xi = \int_0^z \frac{(m_A + m_B)g}{T(z')} \, dz'.$$

When the temperature and concentration gradients vary only in the z-direction (defined by $-g$) one obtains from (5.2)

$$\frac{\partial \ln c}{\partial \xi} = \alpha_{AB} \frac{\partial \ln T}{\partial \xi} + \beta_{AB}, \tag{5.3}$$

where $\alpha_{AB} \equiv (\kappa_A^n - \kappa_A^T)/\kappa_A^c$, and $\beta_{AB} = 3(\kappa_A^n/\kappa_A^c)((M_A - M_B)c + M_B)$ accounts for the effects of gravity. In order to study the (pure) effect of inelasticity on segregation, we consider the case $m_A = m_B$ and $\sigma_A = \sigma_B$, corresponding to a system of particles that differ by their coefficients of restitution only. Clearly, when all coefficients of restitution are equal, the concentration, c, should be uniform. Figure 1(*a*) presents plots of α_{AB} as a function of c, for different values of the degrees of inelasticity. When the coefficients of restitution are different from each other (see e.g. the solid line), the system has non-vanishing concentration gradients. Segregation in the presence of a temperature gradient can therefore occur as a consequence of inelasticity alone. For example, when $\varepsilon_{AA} > \varepsilon_{AB} > \varepsilon_{BB}$ (solid line), species A tends to concentrate in the colder region. Interestingly, segregation occurs even in the unphysical limit when $\varepsilon_{AA} = \varepsilon_{BB} = 0$, but ε_{AB} is different from zero (and the concentrations are different), as shown in figure 1(*a*). This is because the species in excess experiences more inelastic collisions than the other species. Figure 1(*b*) presents results of an MD simulation of a vertically vibrated granular mixture, which demonstrates segregation when the degrees of inelasticity are different from each other but the masses and radii are uniform.

5.2. *Open vibrated binary mixture under gravity: direct and reverse buoyancy*

Consider a roofless vertically vibrated binary mixture under gravity, where energy is supplied at the floor of the system. In the simple case when there is no convection and the system is infinite and homogeneous in horizontal planes, the equations of
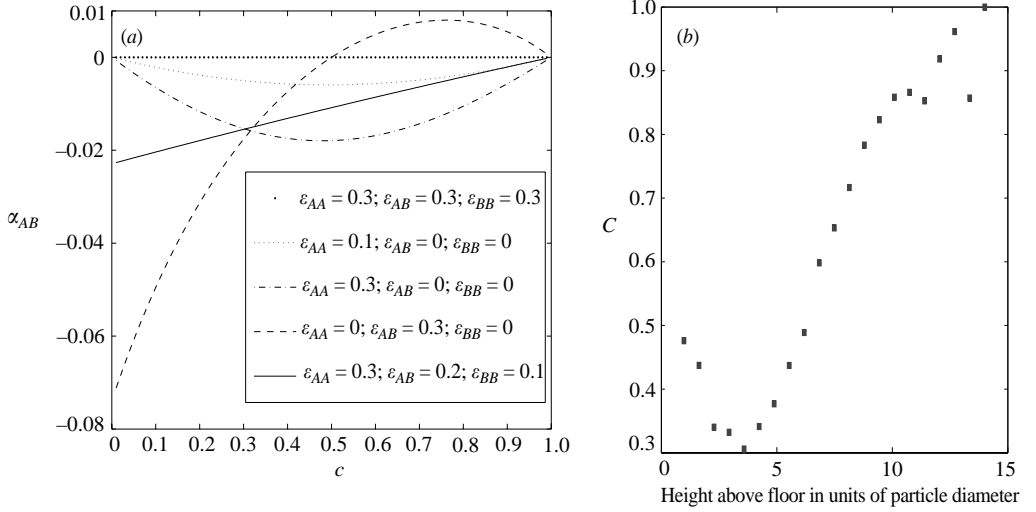
FIGURE 1. (*a*) Plots of the thermal coefficient, α_{AB}, see (5.3), as a function of the concentration, $c \equiv n_A/n$, for different values of the degrees of inelasticity, as detailed in the inset. Here $m_A = m_B$ and $\sigma_A = \sigma_B$. Note that $\alpha_{AB} \neq 0$ when the coefficients of restitution are different. As expected $\alpha_{AB} = 0$ when the coefficients of restitution are all equal, since then the system is monodisperse. (*b*) MD results for the concentration, c, as a function of height in a vertically vibrated system under gravity. The coefficients of normal restitution are: $e_{AB} = 0.83667$, $e_{AA} = 0.94864$ and $e_{BB} = 0.70711$. The system consists of 15625 particles of unit mass and uniform radii equal to 0.0115176, of which 7812 belong to species A. They reside in a container whose floor is a unit square and whose height (which the grains never reach) is 3. The boundary conditions are periodic in the lateral directions. The gravitational acceleration is set to unity. The amplitude and period of vibration of the floor are 0.0075 and 0.075 respectively. The initial condition is a homogeneous mixture of the two species ($c = 0.5$) evenly distributed in a unit cube above the floor.

motion reduce to:

$$\boldsymbol{J}_A = 0, \quad \frac{\mathrm{d}p}{\mathrm{d}z} = -\rho g, \text{ and } -\frac{\partial Q_z}{\partial z} = \Gamma.$$

Define a dimensionless sink term, $\widehat{\Gamma}$ by

$$\Gamma = \frac{n^2 \sigma_{AB}^2}{\sqrt{m_0}} T^{3/2} \widehat{\Gamma}.$$

Using the z-component of (5.1) and (5.2), (4.19), and the equation of motion for the temperature field, one obtains

$$\frac{5\sqrt{6}}{18} \frac{1}{\sigma_{AB}^2 \sqrt{m_0}} \frac{\partial}{\partial z} \left[\sqrt{T} \left(\frac{F_7}{6(M_A c + M_B(1-c))} \frac{\mathrm{d}T}{\mathrm{d}z} + F_8 m_0 g \right) \right] = -\frac{n^2 \sigma_{AB}^2}{\sqrt{m_0}} T^{3/2} \widehat{\Gamma} \quad (5.4)$$

where we recall that $m_0 \equiv m_A + m_B$, and

$$F_7 \equiv 6(\lambda^T - \lambda^n + \lambda^c \alpha_{AB})(M_A c + M_B(1-c)), \quad (5.5)$$

$$F_8 \equiv 3 \left(\lambda^c \frac{\kappa_A^n}{\kappa_A^c} - \lambda^n \right) (M_A c + M_B(1-c)), \quad (5.6)$$

where $M_B = 1 - M_A$ (recall that $M_A \equiv m_A/(m_A + m_B)$). Upon measuring height in terms of the rescaled local pressure $\widehat{p} = (\sigma_{AB}^2/m_0 g)p$, and defining $\eta \equiv -\widehat{p}\partial \ln \sqrt{T}/\partial \widehat{p}$,
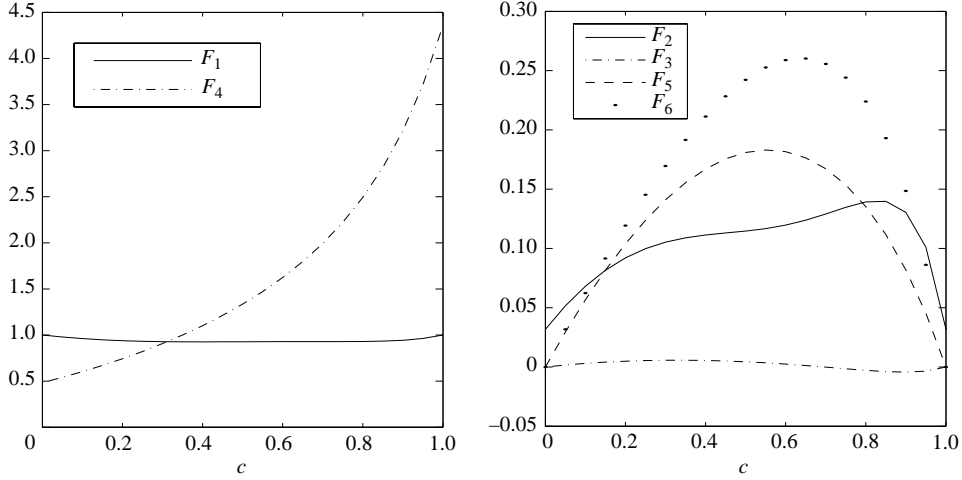
FIGURE 2. The functions $\{F_i\}$ versus the concentration, c, for a mass ratio of 3, equal diameters, and $\varepsilon = 0.1$ for all collisions; see the text for definitions.

(5.2) and (5.4) reduce to the following two first-order coupled ODEs

$$\frac{\partial \eta}{\partial \widehat{p}} = F_1 \frac{\eta^2}{\widehat{p}} - F_2 \frac{\eta}{\widehat{p}} + \frac{F_3}{\widehat{p}} - F_4 \widehat{p}, \quad \frac{\partial c}{\partial \widehat{p}} = -F_5 \frac{\eta}{\widehat{p}} - \frac{F_6}{\widehat{p}}, \tag{5.7}$$

where

$$F_5 \equiv 2\left(\frac{\kappa_A^n - \kappa_A^T}{\kappa_A^c}\right)c, \quad F_6 \equiv \frac{\kappa_A^n}{\kappa_A^c}c, \quad F_1 \equiv 1 + \frac{F_7' F_5}{F_7}, \quad F_2 \equiv -\frac{F_7' F_6}{F_7} - \frac{F_8' F_5}{F_7} - \frac{F_8}{F_7},$$

$$F_3 \equiv \frac{F_8' F_6}{F_7} \text{ and } F_4 \equiv -\frac{9\sqrt{6}}{5}\frac{\widehat{\Gamma}}{F_7((M_A - M_B)c + M_B)}, \text{ and } F_i' \equiv \frac{\partial F_i}{\partial c}.$$

All $\{F_i\}$ are functions of the set of dimensionless parameters, $\mathscr{S}$. The dependence of the functions, $\{F_i(\mathscr{S})\}$, on the concentration, c, is depicted in figure 2 for the case $\varepsilon_{\alpha\beta} = 0.1$, $M_A = 0.25$ and $\sigma_A/\sigma_B = 1$.

We have verified that the functions, $\{F_i(\mathscr{S})\}$, satisfy (see also figure 2) the following properties (as a function of the concentration, c, when all other parameters are kept fixed): (i) All $\{F_i\}$ are bounded; (ii) $F_4 > 0$, $F_5 \geqslant 0$ and $F_6 \geqslant 0$; (iii) $F_3 = F_5 = F_6 = 0$ at $c = 0$ and $c = 1$; (iv) $F_1 = 1$ at $c = 0$ and $c = 1$; (v) $F_5'(c = 0) > 0$, $F_6'(c = 0) > 0$, $F_5'(c = 1) < 0$, $F_6'(c = 1) < 0$, and (vi) $F_1(F_6/F_5)^2 + F_2(F_6/F_5) + F_3 \neq 0$, except at $c = 0$ or $c = 1$. An analysis of (5.7) which employs the above properties of $\{F_i\}$, reveals that the concentration tends to unity at $\widehat{p} = 0$ (corresponding to 'infinite' height above the floor; this is the effect of buoyancy) as well as for $\widehat{p} \to \infty$ (corresponding to an infinite mass above the floor), and has a single minimum at a finite value of $\widehat{p}$. Also, the heat flux at infinite height ($\widehat{p} = 0$) automatically vanishes, as it is supposed to (Brey *et al.* 2001). As (5.7) are first-order coupled ODEs their solution depends on the values of c and η at a given value of $\widehat{p}$ (alternatively, on the average value of c and the heat flux at the floor). The solution also depends on the parameters that enter (5.7), i.e. the mass ratio, the diameter ratio and the coefficients of restitution, but not on the total number of particles. The latter is determined by the choice of the maximal value of $\widehat{p}$ (the minimal being zero, corresponding to infinite height), i.e. the same graph (for $0 \leqslant \widehat{p} \leqslant \infty$) corresponds to any number of particles in the system, each
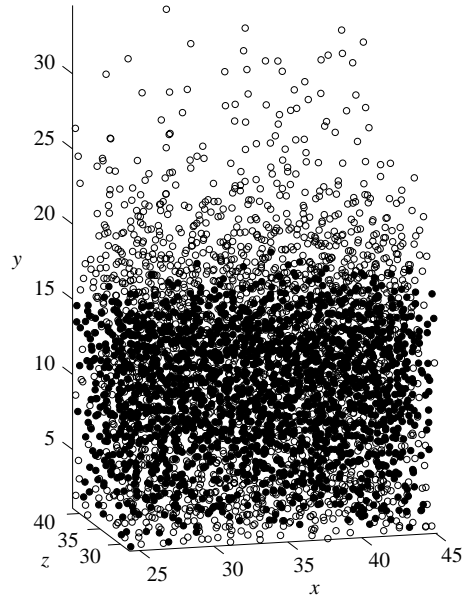
FIGURE 3. Particle scatter plot, based on an MD simulation of a vertically vibrated system under gravity, comprising 32000 particles of unit mass (open circles) and 32000 particles of mass 3 (solid circles). For all collisions $\varepsilon = 0.1$ and all particle radii equal 0.0071985. The base of the computational box is a unit square and its ceiling (which the particles never reach) is of height 3. The boundary conditions are periodic in the lateral directions. The gravitational acceleration is normalized to unity. The amplitude and period of vibration of the floor are 0.012 and 0.15 respectively. The initial condition is a homogeneous mixture of the two species in a unit cube above the floor. The shown length units are in terms of particle diameters. Segregation into three layers is evident. Only part of the system is shown.

such value corresponding to a chosen end of the range of values of $\hat{p}$ (the pressure at the floor is determined by the total number of particles of each species). In this sense the above result is universal. When the value of $\hat{p}$ corresponding to the minimal value of c is within the above range of $\hat{p}$, the (light particle) concentration is maximal at both the floor and the top of the system. In any case $c = 1$ at 'infinite' height (for $\hat{p} = 0$) as expected by the effect of buoyancy. Therefore these equations predict both a quantitative and qualitative degree of universality, the latter being the 'sandwich' arrangement of the light particles. The above conclusions need to be modified for convective flows, and the experimentally important effects of the sidewalls need to be accounted for as well.

A particle scatter plot which demonstrates the above segregation effect in a vertically vibrated system is presented in figure 3. A quantitative comparison of the theoretical and MD results is presented in figure 4; good agreement is obtained, the difference being probably due to finite density effects which we have not accounted for. Figure 5 presents a favourable comparison of theory with the experimental results of Wildman & Parker (2002). While not quantitatively verified, our results are in qualitative agreement with other simulations of vibrated systems (Krouskop & Talbot 2003) and experiments (Wildman, Huntley & Parker 2001; Wildman & Huntley 2003; Huan *et al.* 2004). A similar picture of segregation (in which the particles also differ in size) was obtained in another experiment (Jiang *et al.* 2003), where the effect is referred to as 'sandwich-like segregation'.
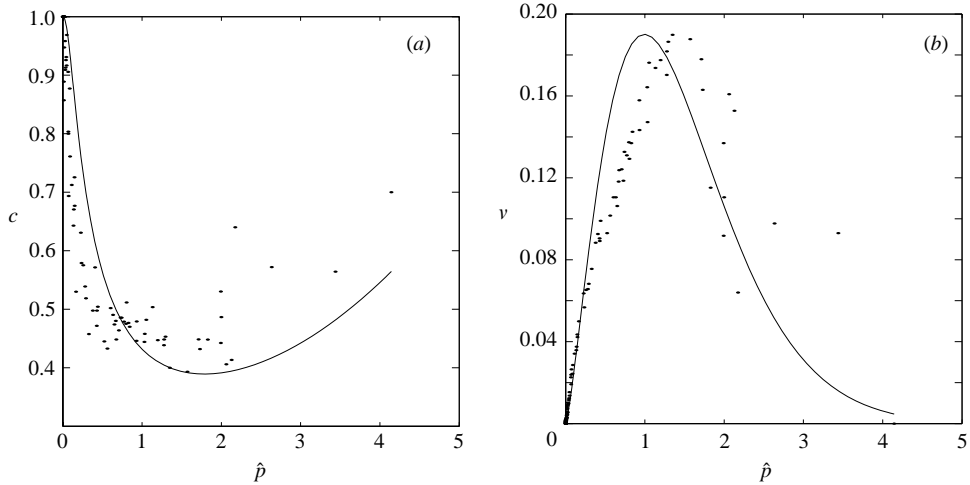
FIGURE 4. (*a*) The concentration, c, versus the dimensionless pressure, $\hat{p}$, for a vertically vibrated system comprising 15625 particles; the mass ratio is 3, the diameter ratio is unity, and all $\varepsilon_{\alpha\beta} = 0.1$. The other parameters are as in figure 1. Smooth lines represent theory and discrete points are results of corresponding MD simulations. (*b*) The volume fraction, ν, versus the dimensionless pressure for the same parameters.
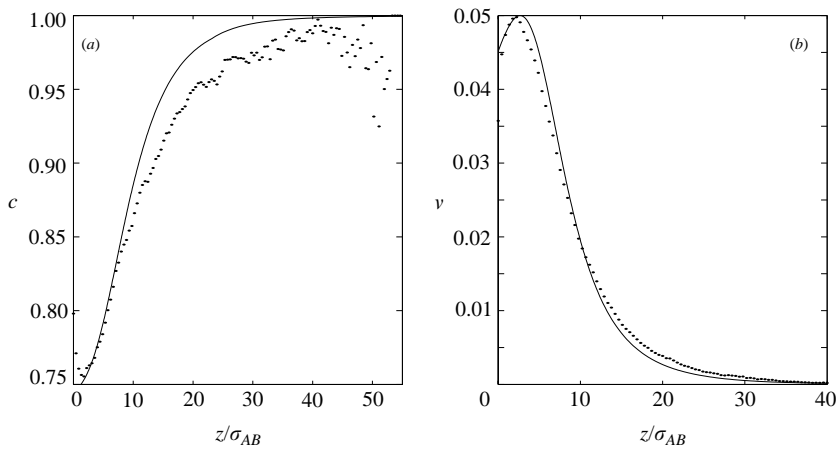

FIGURE 5. (*a*) The concentration, c, versus height in units of the average diameter corresponding to the experiments of Wildman & Parker. Smooth lines represent theory and discrete points correspond to experimental results. The number of A (light) particles is 810 and the number of B (heavy) particles is 175. The radii are $r_A = 2$ mm, $r_B = 2.5$ mm, the particle masses are $m_A = 0.092$ g and $m_B = 0.187$ g, and $e_{AA} = e_{AB} = e_{BB} = 0.91$. The frequency of oscillations is 50 Hz and the amplitude is 1.4 mm (note that the data reported in Wildman & Parker 2002 pertains to an amplitude of 1.74 mm). The slight discrepancy between theory and experiment can be attributed to the effects of the sidewalls, air, finite density effects near the floor, Knudsen effects and possible low statistics at large heights (where hydrodynamics does not apply) or near the floor of the system. (*b*) The volume fraction, ν, versus rescaled height. The other details are the same as for (*a*).

6. Conclusion

The fact that hydrodynamic equations can quantitatively describe fine details of the dynamics of granular gases and granular gas mixtures is rather impressive, given

the lack of scale separation in these gases. The kinetic theory of bidisperse systems is far more complicated than that of monodisperse systems. However, the effort one needs to invest in the study of bidisperse systems is undoubtedly worthwhile, not only because most real granular systems are polydisperse, but because they exhibit a host of interesting phenomena beyond those of the equivalent polydisperse molecular systems. Some of these phenomena, including some novel effects, have been described above, and good agreement with results of simulations and experiments has been demonstrated. A further step is to map out the phase diagram of vibrated granular mixtures. To this end it would help to develop a theory for moderately dense granular mixtures. The shear viscosity in this case is known (Garzo & Montanero 2003). In comparing to experiments one should keep in mind that the effects of air on the grains may be important (cf. e.g. Mobius *et al.* 2005). Although the theory does not signal a 'breakdown' when the ratio of the radii of the constituents is very large, this case needs further study since it corresponds better to a suspension (of the large particles) than a mixture. A derivation of boundary conditions for monodisperse granular gases can be found in Goldhirsch (1999), see also references therein. Boundary conditions for bidisperse systems are yet to be developed. Finally, since, as mentioned, most granular materials are polydisperse, the above theory needs to be further generalized to account for polydispersity, and frightening (to a theoretician) as the prospect may be, for frictional interactions and non-spherical grains.

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