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Abstract. The Fokker–Planck equation for a heavy particle in a granular fluid is derived from the Liouville equation. The host fluid is assumed to be in its homogeneous cooling state and all interactions are idealized as smooth, inelastic hard spheres. The similarities and differences between the Fokker–Planck equation for elastic and inelastic collisions are discussed in detail. Although the fluctuation–dissipation relation is violated and the reference fluid is time-dependent, it is shown that diffusion occurs at long times for a wide class of initial conditions. The results presented here generalize previous results based on the Boltzmann–Lorentz equation to higher densities.
1. Introduction

In the 100 years since Einstein’s remarkable paper on Brownian motion [1], its impact has been felt on a wide range of problems in physics, chemistry and mathematics. The subsequent work of Smoluchowski [2] and Langevin [3] extended his physical model to more precise mathematical structures in the theory of stochastic processes. Attempts to recover these as idealizations of results from the actual Newtonian dynamics of the fluid came much later with the developments of non-equilibrium statistical mechanics [4]. The detailed description of Brownian motion as a function of particle size from first principles remains an open problem [5]. A simpler and more controlled problem is that of a heavy atomic (small size) impurity in an equilibrium host fluid. Since there is a small parameter, the ratio of the host fluid particle mass to that of the impurity (and also small ratio of corresponding mass densities), a systematic analysis would appear to be straightforward. However, even in this case, a simple perturbative analysis is found to be limited in accuracy to its leading asymptotic term and secular terms must be avoided at higher orders [6]. The asymptotic analysis for an impurity in a normal fluid at equilibrium leads to the familiar Fokker–Planck equation [4, 7]. The objective here is to obtain the extension of that result for an impurity in a corresponding granular fluid. These results generalize earlier studies at low density [8, 9].

The idealized prototypical model for a granular fluid is a system of smooth, inelastic hard spheres. This will be the model considered here. The inelasticity of collisions implies a continual loss of energy on each collision. Consequently, the usual equilibrium state for an isolated molecular system is replaced by a homogeneous ‘cooling’ state (HCS), which serves as the environment in which the motion of a heavy impurity is considered. Although this reference HCS is inherently time dependent, an equivalent stationary representation can be given using appropriate velocity scaling. The time dependence of the probability density for the impurity particle in its phase space of position and velocity, can then be analysed by methods similar to those for normal fluids. For example, the general case of diffusion of an impurity particle of arbitrary mass in a fluid undergoing HCS has been given recently in [10]. In this work,
a Zwanzig–Mori projection operator method [11] is used (see appendix A) to obtain a formally
exact kinetic equation valid for this general case. The Fokker–Planck equation then follows from
this kinetic equation to leading order in the ratio of a fluid particle mass to the impurity mass.
The analysis is complicated by the singular nature of the hard sphere interaction, but this mass
ratio expansion has been discussed in detail for hard, elastic collisions [12]. An interesting new
complication for inelastic collisions is a restriction on the cooling rate of the host fluid relative
to the collision frequency of the impurity. If this is too large, the expansion is no longer valid.
This point has already been noted and discussed elsewhere [8, 13].

The rest of the presentation focuses on the similarities and differences between impurity
motion in normal and granular fluids. Firstly, it is shown that there is an exact mapping of the
granular Fokker–Planck equation onto that for elastic collisions, in appropriate dimensionless
variables. Thus, the physical mechanisms of fast velocity relaxation to a Maxwellian for
homogeneous initial states, and fast relaxation to a diffusive equation for inhomogeneous states
occurs for granular gases as well. In particular, the transition to a hydrodynamic stage (diffusion)
takes place for all degrees of inelasticity for the collisions between the impurity and the fluid
particles. However, in terms of physical variables there are significant differences. One of these
is the violation of equipartition, since the kinetic temperature of the impurity is different from
that of the fluid in the stationary state. Also, the usual fluctuation–dissipation relation between
the drift and diffusion coefficients no longer holds, as expected for a non-equilibrium state
of the host fluid. One consequence is that the mobility coefficient, measuring the response to
an external force (for instance, electromagnetic or gravitational), is not simply related to the
diffusion coefficient by the usual Einstein formula.

The Liouville equation for a granular fluid [14, 15] is introduced in the next section. The
special HCS for an isolated system is described and its time dependence is removed through
the introduction of dimensionless variables. It is shown that the HCS is a stationary state for the
Liouville equation in this representation. This stationary representation is used to describe the
motion of an impurity in the fluid. A formally exact kinetic equation is derived for the impurity
particle in section 3, and the condition for its stationary solution is described. Next, in section 4
the form of this kinetic equation is simplified to that of the usual Fokker–Planck equation [16]
in the asymptotic limit of small ratio of fluid mass to impurity mass. The details of the reduction
are given in the appendices. Some of the most important similarities and differences between
impurity motion in normal and granular fluids are listed in section 5, and summary remarks are
given in the last section of the paper.

2. The HCS and its stationary representation

The basis for the analysis to be carried out in this paper is the Liouville equation for a fluid of
$N$ smooth, inelastic hard spheres ($d = 3$) or disks ($d = 2$) of mass $m$ and diameter $\sigma$, and
one impurity of mass $m_0$ and diameter $\sigma_0$. The formal structure of this equation has been
discussed in detail elsewhere in the general context of impurity diffusion [10], and only an
overview will be given here. The position and velocity coordinates of the $N$ equal particles
will be denoted by $\{q_i, v_i; \ i = 1, \ldots, N\}$, while those of the impurity particle by $q_0, v_0$.
The expectation value for some observable $A(\Gamma)$ defined over the $N+1$ particle phase space
$\Gamma \equiv \{q_0, q_1, \ldots, q_N, v_0, v_1, \ldots, v_N\}$, is given by

$$\langle A; t \rangle = \int d\Gamma \rho(\Gamma, t)A(\Gamma), \quad (1)$$
where $\rho(\Gamma, t)$ is the probability density for the state of the system at time $t$ and the dynamics is determined from the Liouville equation:

$$ (\partial_t + \mathcal{L})\rho(\Gamma, t) = 0. \quad (2) $$

The generator for the inelastic hard particle dynamics is

$$ \mathcal{L} = \mathcal{L}_f + v_0 \cdot \nabla_0 - \sum_{i=1}^{N} \tau_{-}(0, i), \quad \mathcal{L}_f = \sum_{i=1}^{N} v_i \cdot \nabla_i - \frac{1}{2} \sum_{i=1}^{N} \sum_{j \neq i}^{N} \tau_{-}(i, j). \quad (3) $$

Here, $\mathcal{L}_f$ is the generator for the dynamics of the fluid particles alone and the subscripts or labels 0 refer to the impurity particle. The second term in the expression of $\mathcal{L}$ generates the free streaming of the impurity. The operators $\tau_{-}(i, j)$ and $\tau_{-}(0, i)$ describe binary scattering between fluid particles and between the impurity and fluid particles, respectively,

$$ \tau_{-}(i, j) = \sigma^{d-1} \int d\Omega |g_{ij} \cdot \sigma| g_{ij} \cdot \sigma |[\alpha^{-2}\delta(q_{ij} - \sigma)b_{ij}^{-1} - \delta(q_{ij} + \sigma)], \quad (4) $$

$$ \tau_{-}(0, i) = \bar{\sigma}^{d-1} \int d\Omega |g_{0i} \cdot \sigma| g_{0i} \cdot \sigma |[\alpha_0^{-2}\delta(q_{0i} - \bar{\sigma})b_{0i}^{-1} - \delta(q_{0i} + \bar{\sigma})], \quad (5) $$

where $d\Omega$ is the solid angle element for the unit vector $\hat{\sigma}$, $\sigma = \sigma \hat{\sigma}$, $\Theta$ is the Heaviside step function, and $g_{ij} = v_i - v_j$ and $q_{ij} = q_i - q_j$ are the relative velocities and positions, respectively. Moreover, $\bar{\sigma} = \bar{\sigma} \hat{\sigma}$ with $\bar{\sigma} = (\sigma + \sigma_0)/2$. The operators $b_{ij}^{-1}$ and $b_{0i}^{-1}$ determine the pre-collision velocities in a restitution collision. They are defined by

$$ b_{ij}^{-1}v_i = v_i - \frac{1 + \alpha}{2\alpha}(g_{ij} \cdot \sigma)\hat{\sigma}, \quad b_{ij}^{-1}v_j = v_j + \frac{1 + \alpha}{2\alpha}(g_{ij} \cdot \sigma)\hat{\sigma}, \quad (6) $$

$$ b_{0i}^{-1}v_0 = v_0 - \frac{(1 + \alpha_0)\Delta}{\alpha_0} (g_{0i} \cdot \sigma)\hat{\sigma}, \quad b_{0i}^{-1}v_i = v_i + \frac{(1 + \alpha_0)(1 - \Delta)}{\alpha_0} (g_{0i} \cdot \sigma)\hat{\sigma}. \quad (7) $$

We have introduced $\Delta = m/(m_0 + m)$, that is the small parameter of the system in the Brownian limit to be discussed in the following. In addition, $\alpha$ and $\alpha_0$ are the coefficients of normal restitution for fluid–fluid and impurity–fluid collisions, respectively. These coefficients measure the degree of inelasticity of collisions and take values in the interval $0 < \alpha, \alpha_0 < 1$. Although in real granular systems the coefficients of normal restitution are known to depend on the relative impact velocity [17], here they will be taken as material constants for the sake of simplicity. It is expected that this approximation keeps the main qualitative features of the phenomenon being investigated.

Average kinetic (granular) temperatures for the fluid $T(t)$ and the impurity particle $T_0(t)$ are defined by

$$ T(t) \equiv \frac{1}{Nd} \sum_{i=1}^{N} m(v_i^2; t) \equiv \frac{1}{2} m\tilde{v}^2(t), \quad (8) $$

$$ T_0(t) \equiv \frac{1}{d} m_0(v_0^2; t) \equiv \frac{1}{2} m_0\tilde{v}_0^2(t). \quad (9) $$
The average velocities $\tilde{v}(t)$ and $\tilde{v}_0(t)$ introduced above will be called thermal velocities of the fluid and the impurity, respectively. Since all the collisions are inelastic and the effect of the impurity on the fluid is negligible in the limit of large $N$, $T(t)$ decreases monotonically in time for an isolated system with $\alpha < 1$. More precisely,

$$\partial_t T(t) = -\zeta(t) T(t),$$

(10)

where $\zeta(t)$ is the ‘cooling’ rate of the fluid due to inelastic collisions,

$$\zeta(t) = -\frac{2N}{v^2(t)} \int d\Gamma v^2_1 T_-(1, 2) \rho(\Gamma, t) \geq 0.$$  

(11)

Similarly, the time evolution of $T_0$ is given by

$$\partial_t T_0(t) = -\zeta_0(t) T_0(t)$$

(12)

with

$$\zeta_0(t) = -\frac{2N}{v^2_0(t)} \int d\Gamma v^2_0 T_-(0, 1) \rho(\Gamma, t),$$

(13)

but it cannot be concluded that the impurity temperature is monotonically decreasing, in general. Its behaviour depends, in detail, on its initial value relative to that of the fluid particles.

Since, the fluid is cooling, there is no stationary solution to the Liouville equation for an isolated system. Instead, it is postulated that there is a solution whose time dependence occurs entirely through $T(t)$ and $T_0(t)$, having the scaling property

$$\rho_{hcs}(\Gamma, t) = [\ell \tilde{v}_{0,hcs}(t)]^{-d}[\ell \tilde{v}_{hcs}(t)]^{-Nd} \rho_{hcs}^*(\{q^*_i, v^*_i, q^*_0, v^*_0\}),$$

(14)

with the dimensionless variables defined by

$$q^*_i = q_i/\ell, \quad q^*_0 = q_0/\ell, \quad v^*_i = v_i/\tilde{v}_{hcs}(t), \quad v^*_0 = v_0/\tilde{v}_{0,hcs}(t).$$

(15)

The coordinates have been scaled relative to $\ell \equiv (n\sigma^d)^{-1}$, where $n$ is the number density of particles, so that $\ell$ is proportional to the mean free path. The subscripts $hcs$ on the thermal velocities denote their values calculated for this particular solution. The dimensionless distribution function $\rho_{hcs}^*$ is invariant under space translations and, therefore, $\rho_{hcs}(\Gamma, t)$ represents a spatially homogeneous state of the fluid plus the impurity. The scaling form in equation (14) implies certain constraints on the cooling rates. Let us introduce reduced cooling rates by

$$\xi_{hcs}^* = \frac{\ell \xi_{hcs}(t)}{\tilde{v}_{hcs}(t)}, \quad \xi_{0,hcs}^* [\gamma(t)] = \frac{\ell \xi_{0,hcs}(t)}{\tilde{v}_{0,hcs}(t)}.$$  

(16)

From equations (11) and (14) it is easily seen that the reduced cooling rate of the fluid $\xi_{hcs}^*$ is time independent. Moreover, the cooling rate for the impurity in this state $\xi_{0,hcs}^*$ depends on time only through the ratio of temperatures

$$\gamma(t) \equiv \frac{\tilde{v}_{0,hcs}(t)}{\tilde{v}_{hcs}(t)} = \sqrt{\frac{T_0(t)m}{T(t)m_0}}.$$  

(17)
Similarly, it is found that the Liouville equation for $\rho^*_{\text{hcs}}$ in these dimensionless variables depends explicitly on time only through $\gamma(t)$. Since $\rho^*_{\text{hcs}}$ is time independent, solutions exist only if $\gamma(t)$ is a constant. The cooling rate equations (10) and (12) then give immediately the requirement

$$
\zeta^*_{\text{hcs}} = \gamma \zeta^*_{0,\text{hcs}}(\gamma).
$$

(18)

In the particular state we are considering, the fluid particles and the impurity particle cool at the same rate, i.e. $\zeta_{\text{hcs}}(t) = \zeta_{0,\text{hcs}}(t)$. This should be viewed as a condition for the existence itself of this state as it fixes one of the two temperatures in terms of the other. However, this does not imply of course that the two temperatures are the same. In fact, it is found that they must be different except in the case of mechanically identical particles, as discussed below. With these results, the equation for $\rho^*_{\text{hcs}}$ becomes independent of time and has the form:

$$
\frac{1}{2} \zeta^*_{\text{hcs}} \sum_{i=0}^{N} \frac{\partial}{\partial v_i} \cdot (v_i^* \rho^*_{\text{hcs}}) + \overline{L}^*(\gamma) \rho^*_{\text{hcs}} = 0,
$$

(19)

where $\overline{L}^*$ is the Liouville operator in dimensionless form (see below). The self-consistent solution to the coupled set of equations formed by equation (19) and the cooling equations determines the HCS. It is the analogue of the Gibbs state for molecular systems and reduces to it for $\alpha = \alpha_0 = 1$.

In dimensionless form, as defined by $\rho^*_{\text{hcs}}$, the HCS is time independent. This suggests a transformation of the Liouville equation to dimensionless variables such that the HCS is a stationary solution in that representation. This can be formally carried out in the following way. We are going to scale the positions and velocities as given by equations (15), where now $\tilde{v}_{\text{hcs}}(t)$ is defined by

$$
\tilde{v}_{\text{hcs}}(t) = \left(2T_{\text{hcs}}/m\right)^{1/2} \text{ and } T_{\text{hcs}}(t) \text{ is the solution of the equation}
$$

$$
\frac{d}{dt}T_{\text{hcs}}(t) = -\zeta_{\text{hcs}}(t)T_{\text{hcs}}(t),
$$

(20)

with the initial condition $T_{\text{hcs}}(t_0) = T(t_0)$, $t_0$ being some arbitrary time. Of course, since in the general case the system is not in the HCS, it is $T(t) \neq T_{\text{hcs}}(t)$. Moreover, in the scaling $\tilde{v}_{\text{hcs}}(t)$ is chosen as given by $\tilde{v}_{0,\text{hcs}} = \gamma \tilde{v}_{\text{hcs}}(t)$, where $\gamma$ is the time-independent parameter identified by equation (18). The appropriate dimensionless time scale $s$ is proportional to the average accumulated number of collisions for the fluid particles in the reference HCS,

$$
s(t, t_0) = \int_{t_0}^{t} \frac{d\tilde{v}_{\text{hcs}}(t')}{\ell}.
$$

(21)

For a general state whose distribution function is $\rho(\Gamma, t)$, the dimensionless distribution $\rho^*(\Gamma^*, s)$ is defined by

$$
\rho(\Gamma, t) = [\ell \tilde{v}_{0,\text{hcs}}(t)]^{-N} [\ell \tilde{v}_{\text{hcs}}(t)]^{-Nd} \rho^*(\Gamma^*, s),
$$

(22)

where $\Gamma^* \equiv \{q^*_0, \ldots, q^*_N, v^*_0, \ldots, v^*_N\}$. Substitution of this into equation (2) gives the dimensionless Liouville equation

$$
(\partial_s + \overline{L}^*) \rho^*(\Gamma^*, s) = 0,
$$

(23)
with the operator $\mathcal{L}^*$ defined by

$$\mathcal{L}^* \Phi(\Gamma^*) = \left[ \gamma \psi_0^* \cdot \nabla_0^* - \gamma \sum_{i=1}^{N} \mathcal{T}^*_{-}(\gamma; 0, i) + \frac{1}{2} \frac{\psi_{hcs}^*}{\psi_0^*} \sum_{i=1}^{N} \frac{\partial}{\partial v_i^*} \cdot v_i^* + \mathcal{T}^*_f + \frac{1}{2} \psi_{hcs}^* \sum_{i=1}^{N} \frac{\partial}{\partial v_i^*} \cdot v_i^* \right] \Phi(\Gamma^*),$$

(24)

for arbitrary $\Phi(\Gamma^*)$. The detailed form of the dimensionless binary collision operators $\mathcal{T}^*_{-}(\gamma; 0, i)$ is given in appendix A. The first three terms on the right-hand side of equation (24) describe the dynamics of the impurity particle. Since the derivative with respect to $s$ in (23) is taken at constant $v_0^*$ and $v_i^*$, some effects of cooling become explicit in the terms proportional to the cooling rate. The last two terms on the right-hand-side of (24) generate the dynamics of the fluid without the impurity, again with explicit effects of cooling. As a consequence of these cooling terms, it is seen that the consistency condition for a stationary solution is the same as equation (19) for the HCS solution,

$$\mathcal{L}^* \rho^*_{hcs} = 0,$$

(25)

since in equation (19) it is:

$$\mathcal{T}^*_{-}(\gamma) = L^*_f + \gamma \psi_0^* \cdot \nabla_0^* - \gamma \sum_{i=1}^{N} \mathcal{T}^*_{-}(\gamma; 0, i).$$

(26)

Therefore, the dimensionless form of the Liouville equation (equation (24)), will be referred to as the stationary representation since it supports a stationary state and that state is the HCS.

3. Kinetic equation for the impurity particle

Consider now the case of the fluid and the impurity in the HCS. At some instant (taken to be $t = t_0 = 0$) the impurity is observed to be at position $\mathbf{R}_0$ with velocity $\mathbf{V}_0$. This initial state can be represented as

$$\rho(\Gamma, 0) = \frac{\rho^*_{hcs}(\Gamma) \delta(\mathbf{R}_0 - \mathbf{q}_0) \delta(\mathbf{V}_0 - \mathbf{v}_0)}{\int d\Gamma \rho^*_{hcs}(\Gamma) \delta(\mathbf{R}_0 - \mathbf{q}_0) \delta(\mathbf{V}_0 - \mathbf{v}_0)}.$$  

(27)

This is equivalent to

$$\rho^*(\Gamma^*, 0) = \frac{\rho^*_{hcs}(\Gamma^*) \delta(\mathbf{R}_0^* - \mathbf{q}_0^*) \delta(\mathbf{V}_0^* - \mathbf{v}_0^*)}{\int d\Gamma^* \rho^*_{hcs}(\Gamma^*) \delta(\mathbf{R}_0^* - \mathbf{q}_0^*) \delta(\mathbf{V}_0^* - \mathbf{v}_0^*)} \equiv \rho^*_{hcs}(\Gamma^*; X_0^*).$$  

(28)

The second equality introduces a convenient notation for the state in which the impurity variables $X \equiv \{ \mathbf{R}, \mathbf{V} \}$ are sharply defined. In the remainder of this discussion, the stationary representation in the dimensionless units will be used, although for simplicity of notation the asterisks will be omitted. The probability $F(\mathbf{R}, \mathbf{V}, s)$ for the impurity to have position and velocity $\mathbf{R}, \mathbf{V}$, at ‘time’ $s$ is then

$$F(X, s) = \int d\Gamma \delta(X - x_0) \rho(\Gamma, t) = \int d\Gamma \delta(X - x_0) e^{-\mathcal{L}^* t \rho^*_{hcs}(\Gamma); X_0^*},$$  

(29)
where, trivially, \( x_0 \equiv \{ q_0, v_0 \} \). A formally exact kinetic equation for this probability is obtained in appendix A. It has the form

\[
\left[ \frac{\partial}{\partial s} + \gamma V \cdot \frac{\partial}{\partial R} + \gamma \Lambda + \frac{\zeta_{hcS}}{2} \frac{\partial^2}{\partial V \cdot V} \right] F(X, s) + \int_0^s ds' \int dX' M(X, X', s - s') F(X', s') = 0.
\]

(30)

The linear operator \( \Lambda \), defined in equation (A.16) in appendix A, describes the exact short time effect of collisions, including the initial correlations among particles in the HCS. If velocity correlations are neglected, \( \Lambda \) reduces to the Enskog–Lorentz collision operator. The form of the operator \( M(X, X', s) \) is given by equation (A.27). It describes dynamically correlated collisions of the impurity with two or more particles that develop in time. These contributions are vanishingly small at short times and generally negligible at low densities. However, at very high densities, they describe the dominant effects of ‘caging’ as the impurity particle becomes more localized due to collisions.

The stationary HCS, \( F_{hcS}(V) \), is determined from equation (30) by

\[
\frac{\zeta_{hcS}}{2} \frac{\partial}{\partial V \cdot [VF_{hcS}(V)]} + \gamma \Lambda F_{hcS}(V) = 0.
\]

(31)

Use has been made of the property given by equation (A.18), i.e.

\[
\int dX' M(X, X', s) F_{hcS}(V') = 0.
\]

(32)

Thus, the HCS is a stationary point of \( M \) but not of \( \Lambda \). Further comment on this is given in the next section. Also, the particularization for the HCS of the expression for the cooling rate given by equation (13) can be expressed in terms of \( F_{hcS} \) as

\[
\zeta_{0, hcS} = \frac{2W}{d} \int dv_0 v_0^2 \Lambda F_{hcS}(v_0),
\]

(33)

where \( W \) being the volume of the system. The results of this section are still exact and serve as a suitable starting point for the small mass ratio expansions to be discussed in the next section.

4. Fokker–Planck limit

In this section, it will be shown that the kinetic equation (30) reduces to a Fokker–Planck equation for asymptotically small \( m/m_0 \). The analysis proceeds in the following way. Firstly, it is shown in appendix B that the operator \( \Lambda \) becomes in the limit \( m/m_0 \rightarrow 0 \)

\[
\gamma \Lambda F(X, s) = -\Gamma_0 \frac{\partial}{\partial V} \cdot \left( V + \frac{G}{2} \frac{\partial}{\partial V} \right) F(X, s),
\]

(34)

where \( \Gamma_0 \) and \( G \) are collision integrals, independent of time and velocity, given by equations (B.17) and (B.20), respectively,

\[
\Gamma_0 = \frac{1 + \alpha_0}{2} \Delta \left( \frac{\sigma}{\sigma} \right)^{d-1} C_1,
\]

(35)
\[ G = \frac{\gamma^{-2}(1 + \alpha_0)\Delta}{2} C_2, \quad (36) \]

\( C_1 \) and \( C_2 \) being real, positive numbers, of the order of unity. Note that in the limit we are considering, \( \gamma^{-2}\Delta \) is a constant, independent of the mass ratio. For elastic collisions, \( G \to 1 \) and \( \Lambda \) becomes the usual Fokker–Planck operator in dimensionless form. The fact that \( G \neq 1 \) here can be considered as a violation of a fluctuation–dissipation relation.

### 4.1. Impurity HCS

Let us analyse the consequences of this for the impurity HCS. Use of equation (34) into equation (31) gives immediately

\[ \frac{\partial}{\partial V} \left[ (1 - \epsilon) V + V^2 \right] F_{hcs} = 0, \quad (37) \]

where we have introduced the parameter

\[ \epsilon \equiv \frac{\zeta_{hcs}}{2\Gamma_0}. \quad (38) \]

It is seen that the solution to this equation is a Gaussian. The normalization and the definition of the impurity temperature require that

\[ F_{hcs}(V) = \pi^{-d/2}e^{-V^2}. \quad (39) \]

Then equation (37) gives the relation

\[ G = 1 - \epsilon. \quad (40) \]

This allows the determination of the impurity temperature in the HCS in terms of the fluid temperature. The latter is determined from equation (10) using for \( \zeta_{hcs} \) the expression identified below, and has a universal value, depending only on \( \alpha \) and being independent of the initial conditions at long times. The impurity temperature in the HCS is then determined from equation (40) with \( G \) given by equation (36),

\[ T_0(t) = \frac{(1 + \alpha_0) C_2}{2(1 - \epsilon)} T(t), \quad (41) \]

following that \( T_0(t) \neq T(t) \).

A further consequence of the above relationship is the condition \( \epsilon < 1 \), i.e.

\[ \frac{\zeta_{hcs}}{2\Gamma_0} < 1. \quad (42) \]

Thus the small mass ratio limit for granular fluids entails, in addition, a restriction on the inelasticity of the fluid–fluid particle collisions. This follows because \( \zeta_{hcs} \propto 1 - \alpha \) and \( \Gamma_0 \propto \Delta \), so the fluid inelasticity must decrease as the mass ratio goes to zero. Asymptotically, for small...
1 − α it is found that (see appendix C)

$$\zeta_{hcs} \sim 4\pi^{(d-1)/2} \left( 1 - \alpha \right) \chi(\sigma),$$  \hspace{1cm} (43)

where \( \chi(\sigma) \) is the fluid–fluid pair correlation function for particles at contact and \( \Gamma(y) \) is the Euler’s \( \Gamma \) function. Then the condition (42) becomes

$$\frac{4\pi^{(d-1)/2}\chi(\sigma)(1-\alpha)}{\sqrt{2\Gamma(d/2)}C_1 d(1+\alpha_0)\Delta} < 1.$$  \hspace{1cm} (44)

In practice, for any choice of \( \Delta \ll 1 \) the value of \( \alpha \) must satisfy \( 1 - \alpha \ll 1 \), also being consistent with equation (44). Otherwise, the derivation of the Fokker–Planck limit for \( \Lambda \) does not hold. It must be realized that this implies that the time for the clustering instability [19] is suppressed. The correct limit for \( \epsilon > 1 \) is described in [13]. Equation (44) can be made more explicit in the low density limit of the fluid. Then, the correlation function \( g_{0}^{(2)} \), appearing in the expression of \( C_1 \), equation (B.18), can be set equal to unity, and \( g_{1}^{(2)} \) can be neglected. In this way, we obtain

$$\frac{1 - \alpha}{\sqrt{2(1+\alpha_0)\Delta}} \left( \frac{\sigma}{\sigma} \right)^{d-1} < 1,$$  \hspace{1cm} (45)

which agrees with the result derived from the Lorentz–Boltzmann equation reported in [8].

In summary, the stationary state condition fixes the form of \( \Lambda \) as a Fokker–Planck operator with friction constant \( \Gamma_0 \) and diffusion coefficient \( (1 - \epsilon) \),

$$\Lambda \mathcal{F} = -\gamma^{-1}\Gamma_0 \frac{\partial}{\partial V} \cdot \left[ V + 1/2(1 - \epsilon) \frac{\partial}{\partial V} \right] \mathcal{F},$$  \hspace{1cm} (46)

where \( \epsilon \) has been defined in equation (36). Moreover, the stationary state equation (37) simplifies to

$$(1 - \epsilon) \frac{\partial}{\partial V} \cdot \left( V + 1/2 \frac{\partial}{\partial V} \right) F_{hcs}(V) = 0.$$  \hspace{1cm} (47)

4.2. Fokker–Planck equation

With the form for \( \Lambda \) given in equation (46), the kinetic equation (30) becomes

$$(\partial_s + \gamma V \cdot \nabla_R) F(X, s) = (1 - \epsilon) \Gamma_0 \frac{\partial}{\partial V} \cdot \left( V + 1/2 \frac{\partial}{\partial V} \right) F(X, s)$$

$$+ \int_0^s ds' \int dX' M(X, X', s - s') F(X', s').$$  \hspace{1cm} (48)

The first term on the right side is now a Fokker–Planck operator of the usual form, but the ‘friction constant’ \( \Gamma_0 \) obtained from \( \Lambda \), has been renormalized by \( (1 - \epsilon) \), due to the additional operator representing cooling.
The values of $\gamma$, $\epsilon$ and $\Gamma_0$ are now known and it remains only to determine the asymptotic form of the last term on the right-hand-side of equation (48). There are two steps in that process. Firstly, since $M(X, X', s')$ involves two binary collision operators it turns out to be proportional to $(\Delta \gamma^{-1})^2$. By exploiting this, in appendix B it is found that

$$\int dX' M(X, X', s - s') F(X', s') \sim \frac{\partial}{\partial \mathbf{V}} \cdot \left( \mathbf{V} + \frac{1}{2} \frac{\partial}{\partial \mathbf{V}} \right) G_c(s - s') F(X, s'),$$

(49)

where $G_c(s)$ is the correlation function

$$G_c(s) = \frac{2}{d} \langle F_+(s) \cdot F_- \rangle_f.$$  

(50)

The brackets denote an equilibrium ensemble average for the fluid in the presence of an infinitely heavy impurity at rest. The phase functions $F_{\pm}$ are the total rate of momentum transfer by the fluid particles to the impurity,

$$F_{\pm} = \gamma^{-1} (1 + \alpha_0^{\pm 1}) \Delta \sigma^{d-1} \sum_{i=1}^N d \Omega (\mp v_i \cdot \hat{\sigma}) \delta(q_{0i} + \mathbf{v}_i \cdot \hat{\sigma}) (v_i \cdot \hat{\sigma})^2 \hat{\sigma}.$$  

(51)

For continuous potentials they would be simply the total force on the impurity, but are determined here for hard spheres by the binary collision operators. The final step in the reduction of the term involving $M(X, X', s - s')$ is to recognize that the time scale for the impurity as described through $F(X, s)$ is slower by a factor $\gamma$ than the decay time for the fluid correlation function $G_c(s)$. To leading order in the mass ratio this gives

$$\int_0^s ds' G_c(s - s') F(X', s') \sim \Gamma_c F(X', s),$$  

(52)

with

$$\Gamma_c = \int_0^\infty ds' G_c(s').$$  

(53)

With all these results, the final form for the Fokker–Planck equation is obtained:

$$(\partial_s + \gamma \mathbf{V} \cdot \nabla_{\mathbf{R}}) F(X, s) = (\Gamma - \epsilon \Gamma_0) \frac{\partial}{\partial \mathbf{V}} \cdot \left( \mathbf{V} + \frac{1}{2} \frac{\partial}{\partial \mathbf{V}} \right) F(X, s),$$  

(54)

where

$$\Gamma = \Gamma_0 + \Gamma_c$$  

(55)

is the total friction constant. In this dimensionless form, the inelasticity of the granular fluid is suppressed and occurs only through the effective friction constant $\Gamma - \epsilon \Gamma_0$.
5. Comparison of granular and normal fluids

The Fokker–Planck equation plays an important role in demonstrating explicitly some important general beliefs of non-equilibrium statistical mechanics. Among these is a two-stage approach to equilibrium. In the present case of a granular fluid, the role of the equilibrium state is played by the HCS. The first stage is a fast relaxation of the velocity distribution on the time scale of a few collisions, followed by a slower transition to spatial homogeneity and full equilibrium by hydrodynamic processes. This follows directly from the Fokker–Planck equation, where the only hydrodynamic process is diffusion.

The recovery of the usual Fokker–Planck equation in dimensionless variables, equation (54), means that many of these qualitative concepts of Brownian motion for normal fluids can be transferred to granular fluids as well. The general solution (Green’s function) for the Fokker–Planck equation can be constructed to demonstrate the two stage relaxation. This has been discussed in some detail for an analysis based on the low-density Boltzmann–Lorentz equation [10] and the analysis there applies here as well. The main similarities between normal and granular fluids are:

- There is a universal stationary state, Gaussian in the velocity and spatially uniform, approached at long times for all physically relevant initial conditions. For initial homogeneous states, the approach to this stationary state is exponentially fast in the time scale $s$ determined by the collision number.

- The spectrum of the Fokker–Planck equation includes an isolated point representing diffusion. This is the smallest point in the spectrum, corresponding to the slowest possible excitation. The next slowest mode decays exponentially fast in $s$ relative to the diffusion mode. Therefore, for general initial conditions, the diffusion equation dominates for sufficiently long times. The existence of diffusion and its dominance applies for all values of the restitution coefficient $\alpha_0$.

This result is relevant because the validity of a hydrodynamic description for granular fluids is not self-evident. Furthermore, if that validity is granted based on empirical or phenomenological grounds, it is often assumed to be limited to weak inelasticity. The Fokker–Planck limit considered here provides an unambiguous example of the existence of hydrodynamics even for strong inelasticity and a description of the initial transient period leading up to its dominance.

There are also significant differences between normal and granular fluids, which have been suppressed by the use of dimensionless variables. Returning to the original variables, equation (54) takes the form

$$\left( \partial_t + \mathbf{V} \cdot \nabla_{\mathbf{R}} \right) F(X, t) = \frac{\partial}{\partial \mathbf{V}} \cdot \left\{ \Gamma [T(t)] \mathbf{V} + \frac{1}{2} D [T(t)] \frac{\partial}{\partial \mathbf{V}} \right\} F(X, t). \quad (56)$$

The first term of the brackets is known as the ‘drift’ term and describes the deterministic dynamics of the particle. The friction coefficient $\Gamma(t)$ is related with the dimensionless one $\Gamma^*$ appearing in equation (54) by $\Gamma(t) = \tilde{\tau}(t) \Gamma^* / \ell$. The second term represents diffusion in velocity space due to the finite temperature of the host fluid and gives rise to all statistical properties of the dynamics. The velocity diffusion coefficient is given by

$$D [T(t)] = \frac{2\gamma T(t)}{m} \left[ \Gamma [T(t)] - \epsilon \Gamma_0[T(t)] \right]. \quad (57)$$
For Brownian motion in a normal fluid, the corresponding relationship is \( D = \frac{(2T/m_0)}{\Gamma} \). This is known as a fluctuation–dissipation relation (velocity diffusion representing fluctuations, the friction constant \( \Gamma \) representing dissipation). This relation is violated for granular fluids in two ways. Firstly, there is the additional term proportional to \( \epsilon \) due to the cooling of the host fluid. Secondly, the impurity particle temperature \( T_0(t) \) differs from that of the host fluid, as shown in equation (41) for the impurity HCS. A related consequence of cooling and the two different temperatures, is a violation of the Einstein relation between spatial diffusion and mobility. The latter can be determined from the Fokker–Planck equation by adding an applied external force and calculating the response of the average velocity of the impurity particle. The resulting mobility coefficient has no simple relationship to the coefficient of diffusion determined from the mean square displacement of the particle.

An obvious effect of the presence of a time dependent temperature in (56), is a nonlinear change in the relevant time scale. This follows from the fact that the fluid–fluid collision frequency sets the physical time scale and this is proportional to \( \sqrt{T(t)} \). Consequently, the dimensionless collision frequency \( s \) depends logarithmically on \( t \). This renormalization of the time is important for identifying the various excitations. For example, the mean square displacement of the impurity becomes linear in the collision number time scale \( s \), not in \( t \).

6. Discussion

The Fokker–Planck equation derived here has several qualifications related to the small parameter \( m/m_o \). In fact, there are other parameters of the problem that can become large such that their product with the small mass ratio compromises the validity of the expansion. If the size of the impurity becomes large relative to the host fluid particle size, the mass densities of the impurity and fluid particles can become comparable. In fact, this is a common experimental condition. In this case, it has been suggested [5] that the Fokker–Planck equation must be modified to a non-Markovian form, since the time scale separation in the analysis of the correlated collisions operator \( M \) is no longer justified.

A second possibility is that the ratio of the impurity temperature to the fluid particle temperature \( T_0/T \) becomes large. This occurs when the cooling rate for the fluid is larger than the effective impurity–fluid collision rate (violation of the condition (42) above). Since the latter decreases with the mass ratio, this requires that the host fluid particle must be less inelastic. When this is not the case, the derivation given here is not valid and the Fokker–Planck description does not apply. Instead, the impurity particle executes ‘ballistic’ rather than diffusive motion [13].

The hard sphere interactions lead to some important differences from the results for continuous potentials. The collision operator in equation (30) has both an instantaneous contribution \( \Lambda \) and one representing finite time correlated collisions \( M \). The first is analogous to the Boltzmann–Enskog–Lorentz collision operator. Accordingly, there are separate contributions from each of them to the friction coefficient \( \Gamma_0 \) and \( \Gamma_c \), respectively. This is puzzling since for continuous potentials the friction coefficient is given by a single Green–Kubo expression, in terms of the time integral of the force autocorrelation function. However, it has been shown recently that this autocorrelation function develops a singularity for steeply repulsive potentials, leading precisely to the instantaneous contribution \( \Gamma_0 \) for hard spheres [20]. The residual non-singular part is given by the ‘force autocorrelation function’ (50). This is a general feature of the hard sphere interaction and is not related to the inelasticity of the collision.
It is interesting that the operator $M$ describing correlated many-body collisions plays no role in determining the HCS distribution for the impurity particle or its cooling rate in the HCS. On the other hand, it does have an important effect for the dynamics of deviations from the HCS, through the correlated part of the friction constant $\Gamma_c$. However, the final temperature of the HCS is independent of this constant.

Within the context described above, the results are exact and apply for both dense and dilute fluids. Explicit evaluation of the friction coefficient as a function of the density and restitution coefficients is a difficult many-body problem. The contribution $\Gamma_0$ is similar to that from the Enskog kinetic theory and in fact reduces to the latter for elastic collisions. Here, it is necessary to understand the pair velocity correlations in the HCS before further simplification is possible. The contribution from correlated binary collisions $\Gamma_c$ is more complicated. Even at low density and elastic collisions, an infinite sequence of ‘ring’ and ‘repeated ring’ recollisions between fluid and impurity particles must be calculated when the size of the impurity becomes comparable to the mean free path [21].

Certainly, the motion of an impurity in a granular fluid provides conceptual, computational and experimental challenges for the second century after Einstein’s initial insight. In this context, we note the recent work by D’Anna et al [18], where experimental results for the motion of a torsion pendulum in a vibrated granular medium is studied. In contrast to the results obtained in this paper, the authors conclude that the system verifies a fluctuation–dissipation relation, similarly to the case of equilibrium molecular fluids. Although the theory developed here is not directly applicable to this case, it can be in principle be analysed by using the same kind of techniques.

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Appendix A. Formal kinetic equation

The projection operator method for normal fluids provides a means to write formally exact equations for observables of interest in terms of collision kernels. These kernels are then the appropriate objects to study by means of chosen approximation schemes [11]. In this appendix, this method is used to obtain the exact kinetic equation for the probability distribution of the impurity particle, defined in equation (29). The method has been discussed in detail in many places for normal fluids and extends directly to the granular fluid when working in the stationary representation. Only a short summary is given here.

The projection operator over the ‘relevant’ part is defined by

$$P \Phi(\Gamma) = \int dX \rho_{hcs}(\Gamma; X) \int d\Gamma' \delta(X-x_0) \Phi(\Gamma'),$$

(A.1)

for an arbitrary phase function $\Phi(\Gamma)$. The distribution function $\rho_{hcs}(\Gamma; X)$ is defined in equation (28). It is easily verified that $P$ actually has the projection property $P^2 = P$. Moreover, it is

$$\int d\Gamma \delta(X-x_0) P \rho(\Gamma, s) = \int d\Gamma \delta(X-x_0) \rho(\Gamma, s) = F(X, s).$$

(A.2)
Consequently, $P\rho(\Gamma, t)$ retains the relevant information in order to evaluate the probability distribution for the impurity. To obtain an equation for $P\rho(\Gamma, t)$, we start from the Liouville equation (23), where the binary collision operator for collisions between the impurity and the fluid particles is

$$
\overline{T}_-(\gamma; 0, i) = \sigma^{d-1} \int d\Omega \left[ \overline{g}_{0i}(\gamma) \cdot \hat{\sigma} \right] \left[ g_{0i}(\gamma) \cdot \hat{\sigma} \right] \left[ \alpha_0^{-2} \delta(q_{0i} - \sigma) \overline{b}_{0i}^{-1} - \delta(q_{0i} + \sigma) \right],
$$

(A.3)

where

$$
\overline{g}_{0i}(\gamma) = v_0 - \gamma^{-1} v_1
$$

(A.4)

and the operator $\overline{b}_{0i}^{-1}$ is defined by

$$
\overline{b}_{0i}^{-1} v_0 = v_0 - \left( 1 + \alpha_0 \right) \Delta \left[ \overline{g}_{0i}(\gamma) \cdot \hat{\sigma} \right] \hat{\sigma},
$$

(A.5)

$$
\overline{b}_{0i}^{-1} v_1 = v_1 + \left( 1 + \alpha_0 \right) \left( 1 - \Delta \right) \gamma \left[ \overline{g}_{0i}(\gamma) \cdot \hat{\sigma} \right] \hat{\sigma}.
$$

(A.6)

We operate on both sides of the Liouville equation with the operator $P$ and rearrange the result to get

$$
(\partial_s + P \overline{L} P) P\rho(\Gamma, s) = -Q \overline{L} Q P\rho(\Gamma; s),
$$

(A.7)

with $Q \equiv 1 - P$. Repeat this analysis by operating on the Liouville equation with $Q$ and rearranging in a similar way,

$$
(\partial_s + Q \overline{L} Q) Q\rho(\Gamma, s) = -Q \overline{L} P\rho(\Gamma, s).
$$

(A.8)

Equations (A.7) and (A.8) are a coupled pair of equations for $P\rho$ and $Q\rho$. They are easily solved with the initial conditions $P\rho(\Gamma; 0) = \rho(\Gamma, 0) = \rho_{hcs}(\Gamma; X_0)$ and $Q\rho(\Gamma; 0) = 0$, following from equation (28), with the result

$$
(\partial_s + P \overline{L} P) P\rho(\Gamma, s) - \int_0^s ds' P \overline{L} e^{-(s-s')Q \overline{L} Q} Q \overline{L} P \rho(\Gamma, s') = 0.
$$

(A.9)

Finally, making some of the projection operators explicit and using equation (A.2), gives the desired kinetic equation

$$
\partial_s F(X, s) + \int dX' B(X, X') F(X', s) + \int_0^s ds' \int dX' M(X, X', s - s') F(X', s') = 0,
$$

(A.10)

with

$$
B(X, X') \equiv \int d\Gamma \delta(X - x_0) \overline{L} \rho_{hcs}(\Gamma; X'),
$$

(A.11)

$$
M(X, X', s) \equiv -\int d\Gamma \delta(X - x_0) \overline{L} e^{-sQ \overline{L} Q} \overline{L} \rho_{hcs}(\Gamma; X').
$$

(A.12)
The kernel $B(X, X')$ represents the instantaneous mean field effects of the fluid on the particle. This can be made more transparent by rewriting its expression in the following way:

$$B(X, X') = \int d\Gamma \delta(X-x_0) \mathcal{L} \delta(X' - x_0) \rho_{hcs}(\Gamma) F^{-1}_{hcs}(V'),$$  \hspace{1cm} (A.13)

where

$$F_{hcs}(V) = \int d\Gamma \delta(X - x_0) \rho_{hcs}(\Gamma)$$  \hspace{1cm} (A.14)

is the impurity distribution in the HCS. Then, using equation (24), it is found

$$B(X, X') = \gamma \left\{ (V \cdot \nabla_R + \Lambda) \delta(X' - X) + \frac{\xi_{0,hcs}}{2} \frac{\partial}{\partial V} \left[ \mathcal{L} \delta(X - X') \right] \right\},$$  \hspace{1cm} (A.15)

with the linear operator $\Lambda$ defined by

$$\Lambda \delta(X' - X) = - \int dx_1 \mathcal{T}_-(\gamma; X, x_1) \delta(X' - X) F^{(2)}_{hcs}(X, x_1) F^{-1}_{hcs}(V).$$  \hspace{1cm} (A.16)

Here

$$F^{(2)}_{hcs}(X, x_1) = N \int dx_0 \int dx_2 \cdots \int dx_N \delta(x_0 - X) \rho_{hcs}(\Gamma)$$  \hspace{1cm} (A.17)

is the reduced two particle distribution for the impurity and one fluid particle, and the operator $\mathcal{T}_-(\gamma; X, x_1)$ is still defined by equation (A.2), but replacing $x_0$ by $X$. Equation (30) follows by substituting equation (A.15) into equation (A.10). As discussed in the main text, $\Lambda$ is closely related to the linearized Boltzmann–Enskog–Lorentz operator for a granular fluid in dimensionless form.

The kernel $M(X, X', s)$ is somewhat more complex. If the complete system is in the HCS, i.e. $\rho(\Gamma, s) = \rho_{hcs}(\Gamma)$ and, consequently, $F(X, s) = F_{hcs}(V)$, it is

$$\int dX' M(X, X', s) F_{hcs}(V') = 0,$$  \hspace{1cm} (A.18)

for all $s$. This is a consequence of the property

$$\int dX' \mathcal{L}(\Gamma) \rho_{hcs}(\Gamma, X') F_{hcs}(V') = \mathcal{L} \rho_{hcs}(\Gamma) = 0.$$  \hspace{1cm} (A.19)

Simplifications of $M$ occur by writing

$$\mathcal{L} = \mathcal{L}_0 + \mathcal{L}_f - \gamma \sum_{i=1}^N \mathcal{T}_-(\gamma; 0, i),$$  \hspace{1cm} (A.20)

where $\mathcal{L}_0$ and $\mathcal{L}_f$ are the generators for the dynamics of the isolated impurity and the isolated fluid, respectively. Their expressions are directly identified from equation (24).
It is easily verified that
\[ \mathcal{P}(\mathcal{L}_0 + \mathcal{L}_f)\mathcal{Q} = 0, \]
and equation (A.12) thus becomes
\[ M(X, X', s) = \gamma \int d\Gamma \delta(X - x_0) \sum_{i=1}^{N} T_{-}(\gamma'; 0, i) e^{-s\mathcal{Q}\mathcal{Q}} Q\mathcal{L}\rho_{hcs}(\Gamma) \delta(X' - x_0) F_{hcs}^{-1}(V'). \]  

(A.21)

Next, use the identity
\[ \mathcal{L}\rho_{hcs}(\Gamma)\Phi(\Gamma) = [\mathcal{L}\rho_{hcs}(\Gamma)] \Phi(\Gamma) + \rho_{hcs}(\Gamma)\mathcal{L}_{-}\Phi(\Gamma) = \rho_{hcs}(\Gamma)\mathcal{L}_{-}\Phi(\Gamma), \]  

valid for arbitrary \( \Phi(\Gamma) \). The new generator \( \mathcal{L}_{-} \) is defined by
\[ \mathcal{L}_{-}\Phi(\Gamma) = \left[ \gamma v_0 \cdot \nabla_0 - \gamma \sum_{i=1}^{N} T_{-}(\gamma'; 0, i) + \frac{\zeta_{hcs}}{2} v_0 \cdot \frac{\partial}{\partial v_0} + \sum_{i=1}^{N} v_i \cdot \nabla_i \right] \Phi(\Gamma), \]  

with the modified binary collision operators
\[ T_{-}(i, j) = \sigma^{d-1} \int d\Omega \delta(q_{ij} - \sigma) \Theta(\mathbf{g}_{ij} \cdot \hat{\sigma}) |\mathbf{g}_{ij} \cdot \hat{\sigma}| \left(b_{ij}^{-1} - 1\right), \]  

(A.25)
\[ T_{-}(\gamma'; 0, i) = \sigma^{d-1} \int d\Omega \delta(q_{0i} - \sigma) \Theta(\mathbf{g}_{0i} \cdot \hat{\sigma}) |\mathbf{g}_{0i} \cdot \hat{\sigma}| \left(b_{0i}^{-1} - 1\right). \]  

(A.26)

For systems with elastic collisions, \( \mathcal{L}_{-} \) is the generator for the time-reversed dynamics, and it plays a similar role for the HCS time correlation functions of granular systems. The detailed proof of equation (A.24) will be given elsewhere. With this identity, equation (A.22) can be rewritten as
\[ M(X, X', s) = -\gamma^2 \int d\Gamma \delta(X - x_0) \sum_{i=1}^{N} T_{-}(\gamma'; 0, i) e^{-s\mathcal{Q}\mathcal{Q}} Q\mathcal{L}\rho_{hcs}(\Gamma) \times \sum_{j=1}^{N} T_{-}(\gamma; 0, j) \delta(X' - x_0) F_{hcs}^{-1}(V'). \]  

(A.27)

Appendix B. Mass ratio expansion

The expressions for \( \Lambda \) and \( M \) are given in equations (A.16) and (A.27), respectively. With the appropriate choice of \( H(x_0, x_1) \), both have the form
\[ Z \equiv \int dx_0 \delta(X - x_0) \int dx_1 \mathcal{T}_{-}(\gamma'; 0, 1)H(x_0, x_1). \]  

(B.1)
which can be transformed, using the expression of the binary collision operator, into
\[
Z = \int dx_0 \int dx_1 \delta (X - x_0) \sigma^{d-1} \int d\Omega \Theta (\vec{g}_{01} \cdot \hat{\sigma}) |\vec{g}_{01} \cdot \hat{\sigma}| \times \left[ \alpha_0^2 \delta(q_{01} - \sigma) b_{01}^{-1} - \delta(q_{01} + \sigma) \right] H(x_0, x_1)
\]
\[
= \int dx_0 \int dx_1 H(x_0, x_1) \delta (R - r_0) \sigma^{d-1} \int d\Omega \Theta (\vec{g}_{01} \cdot \hat{\sigma}) |\vec{g}_{01} \cdot \hat{\sigma}| \delta(q_{01} + \sigma)
\]
\[
\times \left[ \delta(V - \vec{v}_0) - \delta(V - v_0) \right],
\]
(B.2)

where \( \vec{v}_0 \) is obtained from equation (A.5),
\[
\vec{v}_0 = v_0 - (1 + \alpha_0) \Delta (\vec{g}_{01} \cdot \hat{\sigma}).
\]
(B.3)

In the limit of a very massive impurity, it is \( \Delta \gamma^{-1} << 1 \), and \( \delta(V - \vec{v}_0) \) in equation (B.2) can be expanded to second order yielding
\[
Z \sim \int dx_0 \int dx_1 H(x_0, x_1) \delta (R - r_0) \left[ -A(x_0, x_1) \cdot \frac{\partial}{\partial \vec{v}_0} \delta(V - v_0) + \frac{1}{2} B(x_0, x_1) : \frac{\partial}{\partial \vec{v}_0} \frac{\partial}{\partial \vec{v}_0} \delta(V - v_0) \right]
\]
\[
= \int dx_0 \delta(X - x_0)
\]
\[
\times \int dx_1 \left[ \frac{\partial}{\partial \vec{v}_0} \cdot A(x_0, x_1) + \frac{1}{2} \frac{\partial}{\partial \vec{v}_0} \frac{\partial}{\partial \vec{v}_0} : B(x_0, x_1) \right] H(x_0, x_1),
\]
(B.4)

with the definitions
\[
A_i(x_0, x_1) = (1 + \alpha_0) \Delta \sigma^{d-1} \int d\Omega \Theta (\vec{g}_{01}(\gamma) \cdot \hat{\sigma}) \delta(q_{01} + \sigma) |\vec{g}_{01}(\gamma) \cdot \hat{\sigma}|^2 \sigma_i,
\]
(B.5)
\[
B_{ij}(x_0, x_1) = [(1 + \alpha_0) \Delta]^2 \sigma^{d-1} \int d\Omega \Theta (\vec{g}_{01}(\gamma) \cdot \hat{\sigma}) |\vec{g}_{01}(\gamma) \cdot \hat{\sigma}|^3 \sigma_i \sigma_j.
\]
(B.6)

Comparison of equations (B.1) and (B.4) gives the relation
\[
\int dx_1 \Phi_{-}(\gamma; 0, 1) H(x_0, x_1) \sim \frac{\partial}{\partial \vec{v}_0} \cdot \left[ \int dx_1 A(x_0, x_1) H(x_0, x_1) \right]
\]
\[
+ \frac{1}{2} \frac{\partial}{\partial \vec{v}} \cdot \left[ \int dx_1 B(x_0, x_1) H(x_0, x_1) \right].
\]
(B.7)

It should be noted that this is not the complete expansion in the mass ratio, since there remains a dependence on \( \gamma \).

B.1. Evaluation of \( \Lambda \)

For the particular case of \( \Lambda \), equation (B.7) gives
\[
\Lambda F(X, s) = - \int dx_1 \Phi_{-}(\gamma; X, x_1) F^{(2)}_{hcs}(X, x_1) F^{-1}_{hcs}(V) F(X, s)
\]
\[
\sim - \frac{\partial}{\partial V} \cdot \left[ \bar{A}(X) + \frac{1}{2} \frac{\partial}{\partial V} \cdot \bar{B}(X) \right] F(X, s),
\]
(B.8)
with

$$\bar{A}_i(X) = \int dx_0 \delta(x_0 - X) \int dx_1 A_i(x_0, x_1) F_{hcs}^{(2)}(X, x_1) F_{hcs}^{-1}(V) = (1 + \alpha_0) \Delta \left( \frac{\sigma}{\sigma} \right)^{d-1} \times \int dx_0 \delta(x_0 - X) \int dv_1 \varphi_{hcs}(v_1) \int d\Omega \Theta(\hat{g}_{01} \cdot \hat{\sigma})(\hat{g}_{01} \cdot \hat{\sigma})^2 g^{(2)}(\hat{\sigma}, V, v_1) \hat{\sigma}_i, \quad \text{(B.9)}$$

$$\bar{B}_{ij}(X) = \int dx_0 \delta(x_0 - X) \int dx_1 B_{ij}(x_0, x_1) F_{hcs}^{(2)}(X, x_1) F_{hcs}^{-1}(V)$$

$$= [(1 + \alpha_0) \Delta \left( \frac{\sigma}{\sigma} \right)^{d-1} \int dx_0 \delta(x_0 - X) \int dv_1 \varphi_{hcs}(v_1) \int d\Omega \Theta(\hat{g}_{01} \cdot \hat{\sigma})$$

$$\times (\hat{g}_{01} \cdot \hat{\sigma})^3 g^{(2)}(\hat{\sigma}, V, v_1) \hat{\sigma}_i \hat{\sigma}_j]. \quad \text{(B.10)}$$

In the above expressions, we have introduced \(\varphi_{hcs}(v_1) \equiv n^{-1} f_{hcs}(v_1)\), where \(f_{hcs}(v_1)\) is the one-particle distribution of the fluid in the HCS and the correlation function \(g^{(2)}(\hat{\sigma}, V, v_1)\) between the impurity and the fluid particles, defined through

$$F_{hcs}^{(2)}(\hat{\sigma}, V, v_1) = f_{hcs}(v_1) F_{hcs}(V) g^{(2)}(\hat{\sigma}, V, v_1). \quad \text{(B.11)}$$

Because of the spatial homogeneity and isotropy of the HCS, the correlation function must be a function of the scalars \(\sigma, V, v_1, \hat{V} \cdot \hat{\sigma}, \hat{v}_1 \cdot \hat{\sigma}\), and where the hats indicate unit vectors. Furthermore, the characteristic impurity velocity is a factor \(\gamma\) smaller than the characteristic fluid velocity. Therefore, the correlation function may be expanded to first order as

$$g^{(2)}(\hat{\sigma}, V, v_1) \sim g_0^{(2)}(\sigma, v_1, \hat{v}_1 \cdot \hat{\sigma}) + \gamma V \cdot g_1^{(2)}(\sigma, v_1, \hat{v}_1 \cdot \hat{\sigma}). \quad \text{(B.12)}$$

Now equation (B.9) can be written

$$\bar{A}(X) = \gamma^{-1} \Gamma_0 V, \quad \Gamma_0 = \Gamma_0^{(0)} + \Gamma_0^{(1)}, \quad \text{(B.13)}$$

where

$$\Gamma_0^{(0)} \sim (1 + \alpha_0) \Delta \left( \frac{\sigma}{\sigma} \right)^{d-1} \int dv_1 \varphi_{hcs}(v_1) \int d\Omega \Theta(-v_1 \cdot \hat{\sigma}) g_0^{(2)}(\sigma, v_1, \hat{v}_1 \cdot \hat{\sigma})(\hat{V} \cdot \hat{\sigma})$$

$$\times \left[ \gamma^{-1} V^{-1}(v_1 \cdot \hat{\sigma})^2 - 2(v_1 \cdot \hat{\sigma})(\hat{V} \cdot \hat{\sigma}) \right]. \quad \text{(B.14)}$$

The term of order \(\gamma^{-1}\) is of odd parity in \(v_1\) (change \(v_1 \rightarrow -v_1, \hat{\sigma} \rightarrow -\hat{\sigma}\)) and, therefore, it vanishes when integrated over the isotropic distribution \(\varphi_{hcs}(v_1)\). This leaves the dominant term as

$$\Gamma_0^{(0)} = -2(1 + \alpha_0) \Delta \left( \frac{\sigma}{\sigma} \right)^{d-1} \int dv_1 \varphi_{hcs}(v_1) \int d\Omega \Theta(-v_1 \cdot \hat{\sigma})$$

$$\times (v_1 \cdot \hat{\sigma})(\hat{V} \cdot \hat{\sigma})^2 g_0^{(2)}(\sigma, v_1, \hat{v}_1 \cdot \hat{\sigma}). \quad \text{(B.15)}$$

Similarly, the contribution to \(\Gamma\) from \(g_1^{(2)}\) to leading order is

$$\Gamma_0^{(1)} = (1 + \alpha_0) \Delta \left( \frac{\sigma}{\sigma} \right)^{d-1} \int dv_1 \varphi_{hcs}(v_1) \int d\Omega \Theta(-v_1 \cdot \hat{\sigma})(v_1 \cdot \hat{\sigma})^2 (\hat{V} \cdot \hat{\sigma}) \hat{V} \cdot g_1^{(2)}(\sigma, v_1, \hat{v}_1 \cdot \hat{\sigma}). \quad \text{(B.16)}$$
Combining these gives
\[
\Gamma_0 = \frac{(1 + \alpha_0)\Delta}{2} \left( \frac{\bar{\sigma}}{\sigma} \right)^{d-1} C_1 \rightarrow \frac{m}{m_0} \frac{(1 + \alpha_0)}{2} \left( \frac{\bar{\sigma}}{\sigma} \right)^{d-1} C_1,
\]  
where \( C_1 \) is a pure number,
\[
C_1 = 4 \int d\mathbf{v}_1 \varphi_{\text{hcs}}(\mathbf{v}_1) \int d\Omega \Theta(-\mathbf{v}_1 \cdot \hat{\sigma})(\mathbf{v}_1 \cdot \hat{\sigma})(\hat{V} \cdot \hat{\sigma})
\times \left[ -(\hat{V} \cdot \hat{\sigma})g_0^{(2)}(\sigma, \mathbf{v}_1, \hat{v}_1 \cdot \hat{\sigma}) + (\mathbf{v}_1 \cdot \hat{\sigma})\hat{v}_1 \cdot g_0^{(2)}(\sigma, \mathbf{v}_1, \hat{v}_1 \cdot \hat{\sigma}) \right].
\]  
(B.18)

The analysis of \( B_{ij}(X) \) follows in the same way, with the dominant contribution being
\[
B_{ij}(X) = \gamma^{-1} G \Gamma_0 \delta_{ij},
\]  
(B.19)

where
\[
G = \frac{\gamma^{-2}(1 + \alpha_0)\Delta}{2} C_2
\]  
and \( C_2 \) is another pure number,
\[
C_2 = \frac{4}{C_1 d} \int d\mathbf{v}_1 \varphi_{\text{hcs}}(\mathbf{v}_1) \int d\Omega \Theta(\mathbf{v}_1 \cdot \hat{\sigma})(\mathbf{v}_1 \cdot \hat{\sigma})^3 g_0^{(2)}(\sigma, \mathbf{v}_1, \hat{v}_1 \cdot \hat{\sigma}),
\]  
(B.21)

so that \( G \) is independent of the mass ratio in the Brownian limit we are considering.

**B.2. Evaluation of \( M \)**

To evaluate \( M(X, X', s) \) in the small mass ratio limit, first equation (B.7) is substituted into equation (A.27) to get
\[
M(X, X', s) \sim -\gamma^2 \int d\Gamma \delta(X-x_0) \frac{\partial}{\partial \mathbf{v}_0} \cdot \sum_{i=1}^{N} A(x_0, x_i) e^{-s\mathcal{Q}\mathcal{Q}\rho_{\text{hcs}}(\Gamma)}
\times \sum_{j=1}^{N} T_-(\gamma'; 0, j) \delta(X'-x_0) F_{\text{hcs}}^{-1}(V')
\]
\[
= -\gamma \frac{\partial}{\partial \mathbf{V}} \cdot \int d\Gamma \delta(X-x_0) F_+(\Gamma) e^{-s\mathcal{Q}\mathcal{Q}\rho_{\text{hcs}}(\Gamma)}
\times \sum_{j=1}^{N} T_-(\gamma'; 0, j) \delta(X'-x_0) F_{\text{hcs}}^{-1}(V'),
\]  
(B.22)

where we have defined
\[
F_+(\Gamma) = \gamma \sum_{i=1}^{N} A(x_0, x_i) = \gamma(1 + \alpha_0)\Delta \bar{\sigma}^{d-1} \sum_{i=1}^{N} \int d\Omega \Theta(\mathbf{g}_{0i} \cdot \hat{\sigma}) \delta(\mathbf{q}_{0i} + \hat{\sigma})(\mathbf{g}_{0i} \cdot \hat{\sigma})^2 \hat{\sigma}
\sim \gamma^{-1}(1 + \alpha_0)\Delta \bar{\sigma}^{d-1} \sum_{i=1}^{N} \int d\Omega \Theta(-\mathbf{v}_i \cdot \hat{\sigma}) \delta(\mathbf{q}_{0i} + \hat{\sigma})(\mathbf{v}_i \cdot \hat{\sigma})^2 \hat{\sigma}.
\]  
(B.23)
Upon writing equation (B.22), we have neglected the term proportional to $B$ in equation (B.7), because it is of the order $\gamma^{-3}\Delta^2$, while $A$ is of the order $\gamma^{-2}\Delta$. In the case of $\Lambda$, the situation was different because the leading contribution from $A$ happened to vanish identically.

The action of the operator $T_-(\gamma; 0, j)$ on $\delta(X' - x_0)$ can be calculated to leading order for a similar analysis to the one carried out at the beginning of this section, obtaining

$$T_-(\gamma; 0, j)\delta(X' - x_0) = \sigma^{d - 1} \int d\Omega \delta(q_{0j} - \vec{\sigma})\Theta(\vec{g}_{0j} \cdot \vec{\sigma})|\vec{g}_{0j} \cdot \vec{\sigma}| \left[ \delta(X' - \vec{b}_{0j}^{-1} x_0) - \delta(X' - x_0) \right]$$

$$\sim - \frac{(1 + \alpha_0)\Delta}{\alpha_0} \sigma^{d - 1} \int d\Omega \delta(q_{0j} - \vec{\sigma})\Theta(\vec{g}_{0j} \cdot \vec{\sigma})(\vec{g}_{0j} \cdot \vec{\sigma})^2 \vec{\sigma} \cdot \frac{\partial}{\partial V} \delta(X' - x_0). \quad (B.24)$$

Then,

$$M(X, X', s) \sim - \frac{\partial}{\partial V} \int d\Gamma \delta(X - x_0)F_+(\Gamma) e^{-s\Omega Q} Q\rho_{hcs}(\Gamma) F_-(\Gamma) F^{-1}_{hcs}(V') \cdot \frac{\partial}{\partial V} \delta(X' - x_0), \quad (B.25)$$

with

$$F_-(\Gamma) = \frac{\gamma(1 + \alpha_0)\Delta}{\alpha_0} \sigma^{d - 1} \sum_{i=1}^{N} \int d\Omega \Theta(-\vec{g}_{0i} \cdot \vec{\sigma})\delta(q_{0i} + \vec{\sigma})(\vec{g}_{0i} \cdot \vec{\sigma})^2 \vec{\sigma}$$

$$\sim \frac{\gamma^{-1}(1 + \alpha_0)\Delta}{\alpha_0} \sigma^{d - 1} \sum_{i=1}^{N} \int d\Omega \Theta(\vec{v}_i \cdot \vec{\sigma})\delta(q_{0i} + \vec{\sigma})(\vec{v}_i \cdot \vec{\sigma})^2 \vec{\sigma}. \quad (B.26)$$

Since both $F_+(\Gamma)$ and $F_-(\Gamma)$ are proportional to $\gamma^{-1}\Delta$, $M(X, X', s)$ is of order $(\gamma^{-1}\Delta)^2 \sim m/m_0$. This is the same order in the mass ratio as the leading contribution to $\Lambda$. Therefore, all additional corrections on the mass ratio in equation (B.25) should be neglected by consistency. In particular, this means we can substitute

$$e^{-s\Omega Q} Q = e^{-s\Omega Q'} Q \sim e^{-s\Omega Q'} Q = e^{-s\Omega} Q$$

where $\Omega'$ is the generator for the fluid dynamics, including the interactions of the particles with the impurity at rest. More explicitly,

$$\Omega' = \Omega - \sum_{i=1}^{N} \Omega_-(0, i), \quad (B.28)$$

where $\Omega_-$ is the generator for the isolated fluid and

$$\Omega_-(0, i) = \sigma^{d - 1} \int d\Omega (-\vec{v}_i \cdot \vec{\sigma})|\vec{v}_i \cdot \vec{\sigma}| \left[ \alpha_0^{-2}\delta(q_{0i} - \vec{\sigma})b_i^{-1} - \delta(q_{0i} + \vec{\sigma}) \right]. \quad (B.29)$$

$$b_i^{-1} = \frac{1 + \alpha_0}{\alpha_0}(\vec{v}_i \cdot \vec{\sigma})\vec{\sigma}. \quad (B.30)$$

Since $\Omega'$ does not contain dynamics for the impurity, equation (B.25) can be transformed into

$$M(X, X', s) = - \frac{\partial}{\partial V} \int d\Gamma F_+(\Gamma) e^{-s\Omega/\rho_{hcs}(\Gamma)} F_-(\Gamma) \delta(X - x_0) F^{-1}_{hcs}(V') \cdot \frac{\partial}{\partial V} \delta(X - X'), \quad (B.31)$$

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where the relation \( P \rho_{hcs}(\gamma) F_- (\Gamma) \delta(X' - x_0) = 0 \) has been used. Then,

\[
\int dX' M(X, X', s - s') F(X', s') = \frac{\partial}{\partial V} \cdot G_c(s - s') \cdot \left( V + \frac{1}{2} \frac{\partial}{\partial V} \right) F(X, s'),
\]

(B.32)

with

\[
G_c(s) = 2 \int d\Gamma F_+(\Gamma) e^{-\frac{s}{\Omega}} \rho_{hcs}(\Gamma) F_- (\Gamma) \delta(X - x_0) F_{hcs}^{-1}(V).
\]

(B.33)

Because of the homogeneity and isotropy of the HCS, it is

\[
G_c(s) = G_c(s) l,
\]

(B.34)

where \( l \) is the unit tensor, and

\[
G_c(s) = \frac{2}{d} \int d\Gamma_j \left[ F_+ (\Gamma) \cdot e^{-\frac{s}{\Omega}} \rho_{hcs}(\Gamma_j | X) F_- (\Gamma) \right]_{x_0 = X}.
\]

(B.35)

Here \( \Gamma_j \) is the phase space associated to the fluid particles and

\[
\rho(\Gamma_j | X) = [\rho_{hcs}(\Gamma)]_{x_0 = X} F_{hcs}^{-1}(V)
\]

(B.36)

is the conditional HCS probability distribution of the fluid particles given that the phase space coordinates of the impurity are \( X \equiv \{ R, V \} \). To leading order in the mass ratio, we can neglect the velocity correlations between the fluid particles and the impurity, as already discussed in the evaluation of \( \Lambda \). Then, \( G_c(s) \) becomes independent of \( X \). Moreover, in the limit \( \alpha \to 1 \), \( \rho_{hcs}(\Gamma_j | X) \) can be substituted by its equilibrium form. In this way, equation (B.32) becomes

\[
\int dX' M(X, X', s - s') F(X', s') = \frac{\partial}{\partial V} \cdot \left( V + \frac{1}{2} \frac{\partial}{\partial V} \right) G_c(s - s') F(X, s').
\]

(B.37)

This is the expression (49) quoted in the main text.

Appendix C. Evaluation of the cooling rate of the fluid

The dimensionless cooling rate for fluid in the HCS defined by equations (10) and (16) can be reduced to an integral over the appropriate reduced distribution functions. The details are given in appendix A of [10], with the result

\[
\zeta_{hcs} = \frac{1}{2dn} (1 - \alpha^2) \sigma^{d-1} \int d\mathbf{v}_1 \int d\mathbf{v}_2 \int d\Omega f_{hcs}^{(2)}(\mathbf{v}_1, \mathbf{v}_2, \mathbf{q}_{12} = \sigma) \Theta(-\mathbf{g}_{12} \cdot \hat{\sigma}) |\mathbf{g}_{12} \cdot \hat{\sigma}|^3,
\]

(C.1)

where the fluid two-particle distribution function,

\[
f_{hcs}^{(2)}(x_1, x_2) = N(N - 1) \int dx_0 \int dx_3 \cdots \int dx_N \rho_{hcs}(\Gamma),
\]

(C.2)

has been introduced. Due to the condition \( \epsilon < 1 \) (equation (42)), the restitution coefficient \( \alpha \) for the fluid–fluid particle collisions must approach unit as the mass ratio goes to zero.
Hence,

\[ f^{s(2)}(v_1, v_2, q_{12} = \sigma) \sim \chi(\sigma)\varphi_{hcs}(v_1)\varphi_{hcs}(v_2), \quad \varphi_{hcs}(v_i) = \frac{1}{\pi^{d/2}} e^{-v_i^2}, \quad (C.3) \]

where \( \chi(\sigma) \) is the equilibrium pair correlation function at contact. Then, the integrals in equation (C.1) can be performed, and the leading contribution to the cooling rate becomes

\[ \zeta_{hcs} \sim \frac{4\pi(d-1)/2}{\sqrt{2\Gamma(d/2)d}} (1 - \alpha) \chi(\sigma). \quad (C.4) \]

References

[1] Einstein A 1905 *Ann. Phys.* 17 549
[2] Smoluchowski M 1906 *Ann. Phys.* 21 756
[3] Langevin P 1908 *C. R. Acad. Sci. (Paris)* 146 530
[4] See, for example, Lebowitz J L and Resibois P 1965 *Phys. Rev.* A 139 1101
[5] Bocquet L and Hansen J-P 2000 *Dynamics: Models and Kinetic Methods for Non-equilibrium Many Body Systems* (NATO ASI Series vol 371) ed J Karkheck (Dordrecht: Kluwer) p 1
[6] Cukier R I and Deutch J M 1969 *Phys. Rev.* 177 240
[7] Forster D 1975 *Hydrodynamic Fluctuations, Broken Symmetry, and Correlation Functions* (Reading, MA: Benjamin)
[8] Brey J J, Dufty J W and Santos A 1999 *J. Stat. Phys.* 97 281
[9] Brey J J, Ruiz-Montero M I, Cubero D and Dufty J W 1999 *Phys. Rev.* E 60 7174
[10] Dufty J W, Brey J J and Lutsko L 2002 *Phys. Rev.* E 65 051303
    Lutsko J, Brey J J and Dufty J W 2002 *Phys. Rev.* E 65 051304
[11] Zwanzig R 2001 *Nonequilibrium Statistical Mechanics* (Oxford: Oxford University Press)
[12] Bocquet L, Piasecki J and Hansen J-P 1994 *J. Stat. Phys.* 76 505
[13] Santos A and Dufty J W 2001 *Phys. Rev. Lett.* 86 4823
    Santos A and Dufty J W 2001 *Phys. Rev.* E 64 51305
[14] Brey J J, Dufty J W and Santos A 1997 *J. Stat. Phys.* 87 1051
[15] van Noije T and Ernst M H 2001 *Granular Gases* ed T Pöschel and S Luding (New York: Springer) p 3
[16] McLennan J A 1989 *Introduction to Nonequilibrium Statistical Mechanics* (Englewood Cliffs, NJ: Prentice-Hall)
[17] Brilliantov N Y and Pöschel T 2004 *Kinetic Theory of Granular Fluids* (Oxford: Oxford University Press)
[18] D’Anna G, Mayor P, Barrat A, Loreto V and Nori F 2003 *Nature* 424 909
[19] Goldhirsch I and Zanetti G 1993 *Phys. Rev. Lett.* 70 1619
[20] Dufty J W and Ernst M H 2004 *Mol. Phys.* at press (Preprint cond-mat/0401635)
[21] Dorfman J R, van Beijeren H and McClure C F 1980 *J. Stat. Phys.* 23 35