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Mobility and Diffusion in Granular Fluids

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A recent analysis of Brownian motion in granular media undergoing homogeneous cooling implies a violation of the Einstein relation between the diffusion and mobility coefficients. It is shown here that this violation occurs more generally. The usual method of linear response to an external force is generalized for inelastic collisions, where the reference state at zero applied force is the homogeneous cooling state. Formally exact Green–Kubo type expressions are compared for the mobility and diffusion coefficients of an impurity particle in a granular fluid. The results show that the absence of the Gibbs state, the cooling of the reference state, and the occurrence of different kinetic temperatures for the particle and surrounding fluid are responsible for a violation of the Einstein relation. A quantitative description of the effect over a wide range of densities and restitution coefficients is provided by an approximate evaluation of the associated response functions.

KEY WORDS: Granular gases; Green–Kubo expressions; Mobility and diffusion; Einstein relation.

1. INTRODUCTION

The simplest microscopic model of a granular fluid is one composed of hard spheres with inelastic collisions. The isolated fluid supports a homogeneous cooling state (HCS) which is the analogue of the Gibbs equilibrium state. Recently, the transport properties for small spatial perturbations of this state have been studied in detail for a wide range of values for the restitution coefficient using both the Boltzmann kinetic equation[(1)] and its dense fluid extension, the Enskog equation.[(2)] The results support the view that hydrodynamic concepts for fluids with elastic collisions extend quite naturally to those with inelastic collisions when appropriate modifications

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are made.[3] An interesting special case is that of Brownian motion. A detailed description has been developed from an asymptotic expansion in the mass ratio of the fluid particle relative to a heavy impurity particle, based on the Boltzmann–Lorentz equation.[4] The resulting Fokker–Planck equation admits an exact solution that provides the controlled description for a range of transport phenomena. As expected, the usual detailed balance condition associated with the equilibrium Gibbs state is violated. This is reflected in the surprising property that the kinetic temperatures of the fluid and impurity particles are different, although their cooling rates are the same. The predictions of this asymptotic theory have been confirmed by direct Monte Carlo simulation of the Boltzmann–Lorentz equation and by molecular dynamics simulation.[5] An implication of this analysis, not explicitly noted in ref. 4, is that the response to an external force on the impurity particle leads to a mobility coefficient that violates the Einstein relation[10] between the mobility and diffusion coefficients. The objective here is to elaborate on this qualitative difference between real and granular fluids in a more general context, and to identify the mechanisms behind it.

The strongest form of the Einstein relation for equilibrium states is provided by the exact Green–Kubo expressions for the mobility and diffusion coefficients, obtained using linear response methods. Such methods have been extended to diffusion in granular fluids[6] and, as shown below, they can be used to obtain the mobility coefficient for a granular fluid as well. The derivations are formally exact and therefore do not have a priori limitations on the mass ratio, density, or degree of inelasticity. The Einstein ratio between the diffusion coefficient $D(T(t))$ and the mobility $\mu(T(t))$ is defined by

$$\mathscr{E} = \frac{D(T(t))}{T(t)\,\mu(T(t))}. \tag{1}$$

The time dependence of the temperature reflects the fact the reference HCS is cooling. For elastic collisions $\mathscr{E} = 1$, but otherwise it is shown below to be a time independent function of the restitution coefficients for the impurity–fluid and fluid–fluid collisions, as well as the mass and temperature ratios. An approximation to the response functions, expected to be valid for a wide range of these parameters, is used to give a quantitative description of the effect.

The paper is organized as follows. In Section 2, the main physical source for the violation of the Einstein relation is presented by using a phenomenological Langevin equation. The linear response of an impurity particle in a HCS fluid is described in Section 3 to identify the Green–Kubo

expression for the mobility coefficient. In Section 4, this is compared to the corresponding expression for the diffusion coefficient, derived elsewhere.[6] The Einstein ratio $\mathscr{E}$ is shown to be independent of the cooling rate and is expressed in terms of dimensionless "stationary" state response functions. These time correlation functions are evaluated in a leading order cumulant expansion to provide explicit expressions for the Einstein ratio. The results are illustrated for several values of the relevant parameters. It is observed that the approximation considered agrees with a corresponding analysis based on the Enskog kinetic equation. Finally, the results are discussed in Section 5.

2. PHENOMENOLOGICAL OVERVIEW

Linear response for granular systems is inherently different from the usual theory for normal fluids since the reference state is always a nonequilibrium state. This is manifest for the HCS since it has a time dependence through its cooling. Before proceeding with the exact analysis, it is instructional to see the effect of this time dependence on the Einstein ratio in a simple derivation based on a phenomenological Langevin equation for the impurity particle[10]

$$[\partial_t + \gamma(T(t))]\, m_0 \mathbf{v}_0 = \mathbf{f}(t) + \mathbf{F}. \tag{2}$$

Here, $\gamma(T(t)$ is the friction coefficient with an explicit dependence on the fluid temperature, m_0 and $\mathbf{v}_0$ are the mass and velocity, respectively, of the impurity particle, $\mathbf{F}$ is an external applied force, and $\mathbf{f}$ is a stochastic force representing the random effects of fluid particle–impurity particle collisions. The stochastic force has a vanishing average and is "causal"

$$\langle \mathbf{f}(t) \rangle = 0, \qquad \langle \mathbf{f}(t) \cdot \mathbf{v}_0(t') \rangle = 0, \qquad t > t' \tag{3}$$

The solution to the Langevin equation is

$$\mathbf{v}_0(t) = U(t, t')\, \mathbf{v}_0(t') + \int_{t'}^{t} d\tau\, U(t, \tau)[\mathbf{f}(\tau) + \mathbf{F}] \tag{4}$$

where $U(t, t')$ is the solution to Eq. (2) with $\mathbf{f} = \mathbf{F} = 0$ and $U(t', t') = 1$.

The linear response to the external force is obtained directly by averaging Eq. (4) to identify the mobility $\mu(t)$

$$\langle \mathbf{v}_0(t) \rangle = U(t, 0) \langle \mathbf{v}_0(0) \rangle + \mu(t)\, \mathbf{F}, \qquad \mu(t) = \int_0^t d\tau\, U(t, \tau). \tag{5}$$

Similarly, the diffusion coefficient is determined from

$$D(t) = \tfrac{1}{6}\,\partial_t \langle [\mathbf{r}_0(t) - \mathbf{r}_0(0)]^2 \rangle = \tfrac{1}{3}\int_0^t d\tau \langle \mathbf{v}_0(t)\cdot\mathbf{v}_0(\tau)\rangle, \tag{6}$$

where $\mathbf{r}_0$ denotes the position of the impurity particle. In the absence of the external force, the velocity autocorrelation function can be calculated by taking the scalar product of Eq. (4) with $\mathbf{v}_0(t')$ and averaging to get

$$\begin{aligned}\langle \mathbf{v}_0(t)\cdot\mathbf{v}_0(t')\rangle &= U(t,t')\langle v_0^2(t')\rangle + \int_{t'}^t d\tau\, U(t,\tau)\langle \mathbf{f}(\tau)\cdot\mathbf{v}_0(t')\rangle \\ &= U(t,t')\langle v_0^2(t')\rangle. \end{aligned}\tag{7}$$

The diffusion coefficient is given by

$$D(t) = \frac{1}{3}\int_0^t d\tau\, U(t,\tau)\langle v_0^2(\tau)\rangle = \frac{1}{m_0}\int_0^t d\tau\, U(t,\tau)\, T_0(\tau). \tag{8}$$

In the last equality the kinetic temperature of the impurity particle has been introduced by $T_0(\tau) = m_0\langle v_0^2(\tau)\rangle/3$. For systems with elastic collisions the temperature is a constant in time and equal to the fluid temperature, resulting in the Einstein relation $D(t) = T\mu(t)/m_0$. Clearly, the cooling of the HCS due to inelastic collisions invalidates this relation, since the temperature is changing during the response time and contributes differently to the mobility and diffusion coefficients.

The phenomenological analysis given here in terms of the Langevin equation can be made formally exact for elastic collisions using linear response methods. In detail, the exact Einstein relation also requires properties of the Gibbs ensemble. The parallel analysis for inelastic collisions given in the following sections also is formally exact. The violation of the Einstein relation is due to the fact that the HCS is a nonequilibrium state, both in the sense of a time dependent temperature, leading to the effect in Eq. (8), and also the fact that it is not represented by the Gibbs ensemble.

3. LINEAR RESPONSE AND MOBILITY

Consider a fluid of N identical particles of mass m, diameter σ, and interparticle restitution coefficient α. Initially, an additional impurity particle of mass m_0, diameter σ_0, and interparticle restitution coefficient α_0 is added to the system. The position and velocity coordinates of the impurity

particle are denoted by $\mathbf{r}_0$ and $\mathbf{v}_0$. The Liouville equation for the composite $N+1$ particle distribution function is[7]

$$(\partial_t + \bar{L})\, \rho(\Gamma, t) = 0, \tag{9}$$

where the Liouville operator generating the dynamics is given by

$$\bar{L} = \sum_{i=0}^{N} \mathbf{v}_i \cdot \nabla_i - \sum_{i=1}^{N} \bar{T}(i,0) - \tfrac{1}{2} \sum_{i=1}^{N} \sum_{j \neq i}^{N} \bar{T}(i,j)). \tag{10}$$

The first term on the right side generates free streaming while the second terms describe velocity changes. The binary collision operator $\bar{T}(i,j)$ for fluid particles i and j is

$$\bar{T}(i,j) = \sigma^2 \int d\Omega\, \Theta(\mathbf{g}_{ij} \cdot \hat{\boldsymbol{\sigma}})(\mathbf{g}_{ij} \cdot \hat{\boldsymbol{\sigma}})[\alpha^{-2} \delta(\mathbf{r}_{ij} - \boldsymbol{\sigma})\, b_{ij}^{-1} - \delta(\mathbf{r}_{ij} + \boldsymbol{\sigma})], \tag{11}$$

where $d\Omega$ denotes the solid angle integration for the unit vector $\hat{\boldsymbol{\sigma}}$, $\mathbf{r}_{ij}$ is the relative position vector of the two particles, and the operator b_{ij}^{-1} is a substitution operator, $b_{ij}^{-1} F(\mathbf{g}_{ij}) = F(b_{ij}^{-1} \mathbf{g}_{ij})$, which changes the relative velocity $\mathbf{g}_{ij} = \mathbf{v}_i - \mathbf{v}_j$ into its restituting velocity

$$b_{ij}^{-1} \mathbf{g}_{ij} = \mathbf{g}_{ij} - \frac{(1+\alpha)}{\alpha} \hat{\boldsymbol{\sigma}}(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{ij}). \tag{12}$$

The binary collisions between fluid particle i and the impurity are similar to those for collisions among fluid particles

$$\bar{T}(i,0) = \bar{\sigma}^2 \int d\Omega\, \Theta(\mathbf{g}_{i0} \cdot \hat{\boldsymbol{\sigma}})(\mathbf{g}_{i0} \cdot \hat{\boldsymbol{\sigma}})[\alpha_0^{-2} \delta(\mathbf{r}_{i0} - \bar{\boldsymbol{\sigma}})\, b_{i0}^{-1} - \delta(\mathbf{r}_{i0} + \bar{\boldsymbol{\sigma}})], \tag{13}$$

with $\bar{\sigma} = (\sigma + \sigma_0)/2$ and the operator for impurity–fluid restituting collisions is

$$b_{i0}^{-1} \mathbf{g}_{i0} = \mathbf{g}_{i0} - \frac{(1+\alpha_0)}{\alpha_0} \hat{\boldsymbol{\sigma}}(\mathbf{g}_{i0} \cdot \hat{\boldsymbol{\sigma}}). \tag{14}$$

For an isolated system there is no stationary solution to the Liouville equation corresponding to the spatially homogeneous Gibbs state, due to the inherent time dependence associated with loss of energy in collisions. Instead, there is evidence for a homogeneous scaling solution (HCS) whose time dependence occurs entirely through a scaling of the velocity

$$\rho_{\mathrm{hcs}}(\Gamma, t) = (\ell v(t))^{-3(N+1)} \rho^*_{\mathrm{hcs}}(\{\mathbf{r}_{ij}/\ell, \mathbf{v}_i/v(t)\}) \tag{15}$$

The dimensionless distribution function ρ^*_{hcs} is invariant under space translations, with the coordinates scaled relative to the mean free path $\ell \equiv 1/n\sigma^2$. All time dependence occurs through the mean velocity per particle $v(t)$ defined through the total kinetic energy for the system

$$v^2(t) \equiv \frac{2}{3N}\int d\Gamma\, \rho_{\text{hcs}}(\Gamma, t) \sum_{i=1}^{N} \mathbf{v}_i^2 \equiv \frac{2T(t)}{m}. \tag{16}$$

The second equality defines an associated temperature for the fluid. The time dependence of $v(t)$ and $T(t)$ can be calculated from the Liouville equation (9) and the explicit form of $\bar{L}$

$$\partial_t \ln T(t) = -\zeta(t) = 2\partial_t \ln v(t), \tag{17}$$

where ζ is the "cooling" rate

$$\zeta(t) = (1-\alpha^2)\frac{\sigma^2}{6nv^2(t)}\int d\mathbf{v}_1 \int d\mathbf{v}_2 \int d\Omega\, \Theta(\mathbf{g}_{ij}\cdot\hat{\boldsymbol{\sigma}}) \times (\mathbf{g}_{ij}\cdot\hat{\boldsymbol{\sigma}})^3 f^{(2)}_{\text{hcs}}(\mathbf{v}_1, \mathbf{v}_2, \mathbf{r}_{12} = \boldsymbol{\sigma}; t), \tag{18}$$

$f^{(2)}_{\text{hcs}}$ being the reduced two particle distribution associated with $\rho_{\text{hcs}}(\Gamma, t)$. The contribution from the impurity particle vanishes in the thermodynamic limit and has been neglected. The reduced distribution has the same scaling property as $\rho_{\text{hcs}}(\Gamma, t)$ so it is easily verified that $\zeta(t) \propto v(t)$. Therefore, Eq. (17) can be integrated for the explicit time dependence of $v(t)$ and $T(t)$

$$v(t) = v(0)[1+\tfrac{1}{2}\zeta(0)\, t]^{-1}, \qquad T(t) = T(0)[1+\tfrac{1}{2}\zeta(0)\, t]^{-2}. \tag{19}$$

Substitution of Eq. (17) into the Liouville equation gives

$$\bar{\mathscr{L}}\rho_{\text{hcs}} = 0, \qquad \bar{\mathscr{L}} \equiv \bar{L} + \tfrac{1}{2}\zeta(t)\left[3(N+1) + \sum_{i=0}^{N} \mathbf{v}_i \cdot \nabla_{v_i}\right], \tag{20}$$

where $\nabla_{v_i} \equiv \partial/\partial\mathbf{v}_i$. The self-consistent solution to the coupled set of equations (18) and (20) completely determines ρ_{hcs}. The dependence on the (dimensionless) relative coordinates $\{r_{ij}/\ell\}$ assures that it is translational invariant and represents a spatially homogeneous fluid.

Now consider a perturbation of the HCS due to a small constant force $\mathbf{F}$ exerted only on the impurity particle. The Liouville equation to linear order in this force is

$$(\partial_t + \bar{L})\, \rho(t) = -\frac{\mathbf{F}}{m_0}\cdot\frac{\partial}{\partial\mathbf{v}_0}\rho_{\text{hcs}}(t). \tag{21}$$

Before solving this equation, it is useful to introduce dimensionless variables to eliminate the time dependence of the HCS. An appropriate set of dimensionless particle coordinates, velocities, and time are given by

$$\mathbf{r}_i^* = \mathbf{r}_i/\ell, \qquad \mathbf{r}_{ij}^* = \mathbf{r}_{ij}/\ell, \qquad \mathbf{v}_i^* = \mathbf{v}_i/v(t), \qquad s(t) \equiv \frac{1}{\ell}\int_0^t d\tau\, v(\tau). \tag{22}$$

The dimensionless time scale is the integral of the average collision frequency and thus is a measure of the number of collisions in time t. In terms of these variables, Eq. (21) becomes

$$(\partial_s + \bar{\mathscr{L}}^*)\, \rho^* = -\mathbf{F}^*(s)\cdot\frac{\partial}{\partial \mathbf{v}_0^*}\, \rho_{\text{hcs}}^*, \tag{23}$$

where $\bar{\mathscr{L}}^*$ is the dimensionless form of $\bar{\mathscr{L}}$ in Eq. (20). It has the same form in the dimensionless variables, with $\sigma \to \sigma^*$, $\bar{\sigma} \to \bar{\sigma}^*$ and $\zeta(t) \to \zeta^*$. Here,

$$\sigma^* = \sigma/\ell, \qquad \bar{\sigma}^* = \bar{\sigma}/\ell, \qquad \zeta^* = \ell\zeta(t)/v(t), \qquad \mathbf{F}^*(s) = \frac{\ell \mathbf{F}}{m_0 v^2(t)}, \tag{24}$$

$$\rho(\Gamma, t) = (\ell v(t))^{-3(N+1)}\, \rho^*(\{\mathbf{r}_{ij}^*, \mathbf{v}_i^*\}, s). \tag{25}$$

Note that the generator for the isolated fluid dynamics is independent of s as is ρ_{hcs}^*. However, the external force has inherited a dependence on s due to the velocity and time scaling. The formal solution to Eq. (23) is

$$\rho^*(s) = e^{-(s-s_0)\,\bar{\mathscr{L}}^*}\rho^*(s_0) - \int_{s_0}^{s} ds'\, e^{-(s-s')\,\bar{\mathscr{L}}^*}\frac{\partial \rho_{\text{hcs}}^*}{\partial \mathbf{v}_0^*}\cdot\mathbf{F}^*(s') \tag{26}$$

The average velocity of the particle at "time" s follows directly from an average over the solution (26)

$$\langle \mathbf{v}_0^*; s\rangle = \langle \mathbf{v}_0^*; s_0\rangle + 2\int_{s_0}^{s} ds'\, R_\mu(s-s')\, \mathbf{F}^*(s'), \tag{27}$$

where

$$\langle \mathbf{v}_0^*; s\rangle = \int d\Gamma^*\, \mathbf{v}_0^*\, \rho^*(s) \tag{28}$$

and the response function $R_\mu(s)$ is

$$R_\mu(s) = -\frac{1}{6}\int d\Gamma^*\, \mathbf{v}_0^*\cdot e^{-s\bar{\mathscr{L}}^*}\frac{\partial \rho_{\text{hcs}}^*}{\partial \mathbf{v}_0^*} = -\frac{1}{6}\left\langle \mathbf{v}_0^*(s)\cdot\frac{\partial \ln \rho_{\text{hcs}}^*}{\partial \mathbf{v}_0^*}\right\rangle_{\text{hcs}} \tag{29}$$

The notation here is

$$\mathbf{v}_0^*(s) = e^{s\mathscr{L}^*}\mathbf{v}_0^*, \qquad \langle X\rangle_{\rm hcs} = \int d\Gamma^*\, \rho_{\rm hcs}^* X(\Gamma^*), \tag{30}$$

where $\mathscr{L}^*$ is the adjoint of $\bar{\mathscr{L}}^*$, given explicitly in Appendix A. Restoring the dimensions to the average velocity and force gives

$$\begin{aligned}\langle \mathbf{v}_0; s\rangle &= \langle \mathbf{v}_0; s_0\rangle + \frac{2\ell}{m_0 v(t)}\int_{s_0}^{s} ds'\, \frac{v^2(t)}{v^2(t')}\, R_\mu(s-s')\,\mathbf{F} \\ &= \langle \mathbf{v}_0; s_0\rangle + \frac{2\ell}{m_0 v(t)}\int_{s_0}^{s} ds'\, e^{-\zeta^*(s-s')} R_\mu(s-s')\,\mathbf{F}.\end{aligned} \tag{31}$$

The second equality uses the explicit form $v^2(t) = v^2(0)\, e^{-\zeta^* s}$ as follows from (17), (19), and (24). Finally, consider the case where the force is applied in the remote past when the fluid is in the HCS. Then Eq. (31) can be written as

$$\langle \mathbf{v}_0; s\rangle = \langle \mathbf{v}_0; -\infty\rangle + \mu(T(t))\,\mathbf{F} \tag{32}$$

which identifies the mobility coefficient as

$$\mu(T(t)) = \frac{2\ell}{m_0 v(t)}\int_0^{\infty} ds'\, e^{-\zeta^* s'} R_\mu(s'). \tag{33}$$

There is an explicit time dependence through the temperature, as expected, due to the cooling of the granular fluid. However, there is an additional effect of the cooling during the evolution of the response so that the mobility coefficient is not simply determined by the time integral of the response function. Instead there is a new factor of $e^{-\zeta^* s'}$ multiplying the response function.

As noted in ref. 3, the HCS is unstable to long wavelength perturbations, with the critical wavelength $\lambda_c = 2\pi\sqrt{\eta/2\zeta}$, where η is the shear viscosity. The above limiting form for response at asymptotically late times must be understood in the following context. For systems of largest dimension less than λ_c the system is stable and the formal long time limit is limited only by the Poincare recurrence time. In this case, as for normal fluids a plateau value should be reached after several collisions that is common for all experimentally relevant future times. The technical problem of finite Poincare recurrence time is not relevant for time scales of experiments or simulations, but can be avoided altogether by taking the limit of infinite volume at fixed density. Now there is a time scale for the onset of the instability

that can occur on the relevant time scales for simulation and experiment. Equation (21) assumes the deviation of $\rho(t)$ from $\rho_{\rm hcs}(t)$ is small for sufficiently small $\mathbf{F}$, but this is not the case for times large compared to that for the onset of instability. Thus, the mobility and diffusion coefficients are meaningful only if the time for instability is longer than the few collision times required to reach the plateau value. At low density this is clearly the case, even at strong dissipation, as demonstrated by both Monte Carlo[8] and molecular dynamics simulations.[9] At high densities and large dissipation, it appears that the instability occurs too rapidly for the present analysis based on the HCS to be meaningful.

4. IMPURITY DIFFUSION AND EINSTEIN RELATION

The diffusion of the impurity particle in the HCS has been studied in a similar way using linear response to an initial perturbation of the probability density for the impurity. The details are given elsewhere[6] so only the result is quoted here. The diffusion coefficient D is given by

$$D(T(t)) = \ell v(t) \int_0^\infty ds'\, R_D(s'), \tag{34}$$

where the response function $R_D(s)$ is

$$R_D(s) = \tfrac{1}{3} \langle \mathbf{v}_0^*(s) \cdot \mathbf{v}_0^* \rangle_{\rm hcs}. \tag{35}$$

The Einstein ratio defined in Eq. (1) therefore can be written as

$$\mathscr{E} = \frac{D(T(t))}{T(t)\,\mu(T(t))} = \frac{m_0}{m} \frac{\int_0^\infty ds'\, R_D(s')}{\int_0^\infty ds'\, e^{-\zeta^* s'} R_\mu(s')}. \tag{36}$$

In the elastic limit $\zeta^* \to 0$, $R_\mu(s) \to (m_0/m)\, R_D(s)$, and the usual Einstein ratio $\mathscr{E} \to 1$ is recovered. However, at finite inelasticity the relationship between $D(T(t))$ and $\mu(T(t))$ is no longer simple. There are three separate reasons for this difference. First, the additional factor of $e^{-\zeta^* s'}$ in the integration over the response function for $\mu(T(t))$ arises because the temperature dependence of the mobility and diffusion coefficient are different. Since the temperature is changing during the correlation time for the response, it generates different dynamics in the two cases. The second reason for a change in the Einstein ratio is that $\rho_{\rm hcs}$ is not the Gibbs state and consequently $\partial \ln \rho_{\rm hcs}^*/\partial \mathbf{v}_0^* \neq -2(m_0/m)\, \mathbf{v}_0^*$, i.e., the response function for the mobility is no longer proportional to the velocity autocorrelation function. A third reason, not apparent at this formal level, is that the temperature of

the impurity particle is different from that of the fluid except when the impurity is mechanically equivalent to particles of the fluid.[11]

Equation (36) is still exact and is the primary result of this analysis. To estimate the dependence of $\mathscr{E}$ on the relevant parameters (restitution coefficients, mass ratio, size ratio) the response functions are expanded to leading order in a cumulant expansion[6]

$$R_D(s) \to R_D(0)\, e^{-\omega_D^* s}, \qquad R_\mu(s) \to R_\mu(0)\, e^{-\omega_\mu^* s} \tag{37}$$

with

$$\omega_D^* = -\frac{\langle (\mathscr{L}^* \mathbf{v}_0^*) \cdot \mathbf{v}_0^* \rangle_{\rm hcs}}{\langle v_0^{*2} \rangle_{\rm hcs}}, \tag{38}$$

$$\omega_\mu^* = -\frac{\left\langle (\mathscr{L}^* \mathbf{v}_0^*) \cdot \dfrac{\partial \ln \rho_{\rm hcs}^*}{\partial \mathbf{v}_0^*} \right\rangle_{\rm hcs}}{\left\langle \mathbf{v}_0^* \cdot \dfrac{\partial \ln \rho_{\rm hcs}^*}{\partial \mathbf{v}_0^*} \right\rangle_{\rm hcs}} = \frac{1}{3} \left\langle (\mathscr{L}^* \mathbf{v}_0^*) \cdot \frac{\partial \ln \rho_{\rm hcs}^*}{\partial \mathbf{v}_0^*} \right\rangle_{\rm hcs}. \tag{39}$$

This type of expansion is exact at short times or in the limit of large impurity/fluid mass ratio. For elastic collisions it is known to provide a good approximation for longer times as well, and it is reasonable to expect that this accuracy extends also to inelastic collisions. The mobility and diffusion coefficients become

$$\mu(T(t)) \to \frac{2\ell}{v(t)\, m_0} R_\mu(0) \frac{1}{\omega_\mu^* + \zeta^*} = \frac{\ell}{v(t)\, m_0} \frac{1}{\omega_\mu^* + \zeta^*}, \tag{40}$$

$$D(T(t)) \to v(t)\, R_D(0) \frac{\ell}{\omega_D^*} = \frac{1}{2} \ell v(t) \frac{T_0(t)}{T(t)} \frac{m}{m_0} \frac{1}{\omega_D^*}, \tag{41}$$

where use has been made of the results

$$R_\mu(0) = -\frac{1}{6} \left\langle \mathbf{v}_0^* \cdot \frac{\partial \ln \rho_{\rm hcs}^*}{\partial \mathbf{v}_0^*} \right\rangle_{\rm hcs} = \frac{1}{2}, \tag{42}$$

$$R_D(0) = \frac{1}{3} \langle \mathbf{v}_0^{*2} \rangle_{\rm hcs} = \frac{1}{2} \frac{m}{m_0} \frac{T_0}{T}. \tag{43}$$

Here, it has been recognized that the temperature of the impurity particle $T_0(t)$ is different from that of the fluid for granular systems, as noted above. However, the cooling rates are the same so that $T_0(t)/T(t)$ is time

independent. The specific dependence of $T_0(t)/T(t)$ on restitution coefficients, mass ratio, and size ratio is complex and is discussed in detail in ref. 11. The Einstein ratio becomes

$$\mathscr{E} = \frac{T_0(t)}{T(t)} \frac{\omega_\mu^* + \zeta^*}{\omega_D^*}. \tag{44}$$

The expressions for ω_μ^*, ω_D^*, and ζ^* are evaluated in Appendix B with the results

$$\omega_D^* = -\frac{1}{2}\zeta^* + \nu^* \left[1 + \frac{c_0}{16} \frac{4\theta+3}{(1+\theta)^2} - \frac{c_1}{16} \frac{\theta^2}{(1+\theta)^2} \right], \tag{45}$$

$$\omega_\mu^* = \omega_D^* - \frac{c_0}{4} \frac{\nu^*}{(1+\theta)}, \tag{46}$$

$$\zeta^* = (1-\alpha^2)\, \chi \frac{4}{3} \sqrt{\frac{\pi}{2}} \left(1 + \frac{3}{32} c_1 \right), \tag{47}$$

with the definitions

$$\nu^* = \frac{4}{3} \sqrt{\pi} \frac{m}{m+m_0} \left(\frac{\bar{\sigma}}{\sigma} \right)^2 \chi_0 (1+\alpha_0)\, \theta^{-1/2} (1+\theta)^{1/2}, \tag{48}$$

$$\theta \equiv \frac{m_0}{m} \frac{T(t)}{T_0(t)}. \tag{49}$$

The frequency ν^* is an average impurity–fluid collision frequency, depending on the mass ratio and the impurity–fluid collision restitution coefficient. In contrast, the cooling rate depends only on fluid properties. The constant c_1 measures the deviation of the fluid velocity distribution from Maxwellian and is given by Eq. (B8). Similarly, c_0 measures the deviation of the impurity velocity distribution from Maxwellian. It is more complex and is described in ref. 11. Both c_0 and c_1 vanish in the limit of elastic collisions, and otherwise give contributions of the order of a few percent to ω_μ^*, ω_D^*, and ζ^* even at very strong dissipation. The density dependence of $\mathscr{E}$ occurs only through the factor χ in ζ^* and the factor χ_0 in ω_μ^* and ω_D^*. These are the configurational pair correlation functions for the fluid–fluid and impurity–fluid pairs at contact, respectively. These functions have a density dependence that differs only when the size ratio differs from unity. Thus the Einstein ratio becomes independent of density for same size particles.

Equations (45)–(49) determine the mobility and diffusion coefficients through Eqs. (40) and (41). Approximate expressions for these transport

coefficients can be obtained as well from the Enskog–Lorentz kinetic equation by application of linear response (perturbation by a small external force) for the mobility and by the Chapman–Enskog method for the diffusion coefficient. It is interesting to note that the results of the cumulant expansion agree in detail with these kinetic theory results (first Sonine approximation).[12] In addition, special limits of small fluid–impurity mass ratio[4] and elastic fluid–fluid interactions (but inelastic impurity–fluid)[13] are also recovered. A closely related analysis of self-diffusion has been given by Brilliantov and Pöschel,[14] who also use a cumulant expansion for the velocity autocorrelation function. However, their analysis is in real time for which the velocity autocorrelation function is not stationary. This complication leads them to uncontrolled approximations on the cooling and their resulting self-diffusion coefficient does not agree with that from Boltzmann or Enskog kinetic theory.

A simpler, and still accurate, form for the Einstein ratio is obtained by neglecting the small effects of c_0 and c_1

$$\mathscr{E} \to \frac{T_0}{T}\frac{1+\zeta^*/2\nu^*}{1-\zeta^*/2\nu^*}, \tag{50}$$

$$\frac{\zeta^*}{\nu^*} = \frac{(1-\alpha^2)}{(1+\alpha_0)}\frac{m+m_0}{m}\frac{\chi}{\chi_0}\left(\frac{\sigma}{\bar{\sigma}}\right)^2\sqrt{\frac{\theta}{2(1+\theta)}}. \tag{51}$$

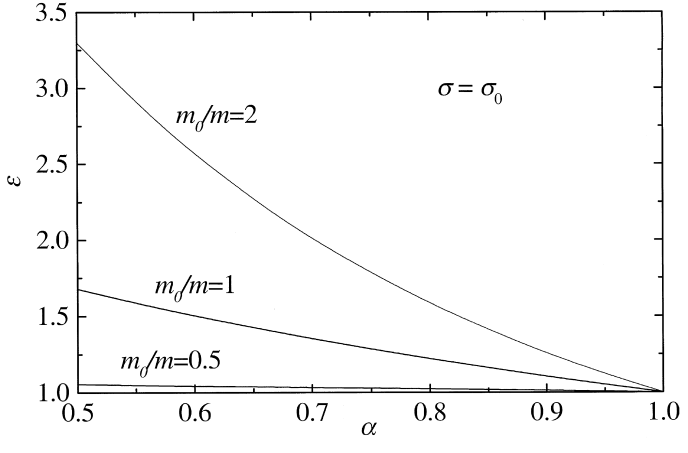

Fig. 1. Plot of the Einstein ratio $\mathscr{E}$ as a function of the restitution coefficient $\alpha = \alpha_0$ for $\sigma = \sigma_0$, and three different values of the mass ratio: $m_0/m = 0.5$, $m_0/m = 1$, and $m_0/m = 4$.

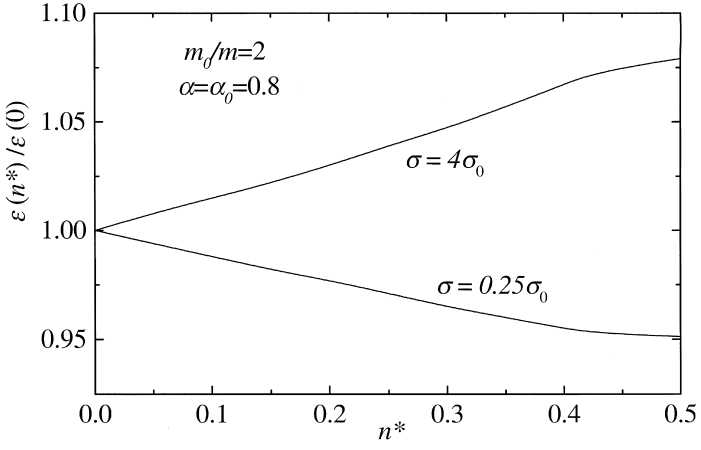

Fig. 2. Plot of the ratio $\mathscr{E}(n^*)/\mathscr{E}(0)$ as a function of the reduced density $n^* = n\sigma^3$ for $\alpha = \alpha_0 = 0.8$, $m_0/m = 2$ and two values of the size ratio: $\sigma/\sigma_0 = 0.25$, and $\sigma/\sigma_0 = 4$.

This shows most directly the dependence on mass ratio, size ratio, restitution coefficients, and temperature differences. For mechanically identical particles this reduces to

$$\mathscr{E} \to \frac{3-\alpha}{1+\alpha}. \tag{52}$$

To illustrate the influence of dissipation on the Einstein ratio $\mathscr{E}$ more generally, consider the particular case $\alpha = \alpha_0$. Figure 1 shows the dependence of $\mathscr{E}$ on the restitution coefficient α for $\sigma = \sigma_0$ and three different values of the mass ratio m_0/m. As might be expected, the deviation of $\mathscr{E}$ from 1 increases with decreasing α, and is greatest when the impurity particle is heavier than the fluid particles. The dependence of $\mathscr{E}$ on the reduced density $n^* = n\sigma^3 = \sigma/\ell$ is shown in Fig. 2 for $m_0/m = 2$, $\alpha = 0.8$, and two different size ratios: $\sigma/\sigma_0 = 0.25$, and 4. In this figure the extended Carnahan–Starling approximation for the pair correlation functions χ and χ_0 as a function of n^* has been used,[15]

$$\chi = \left(1 - \frac{1}{12}\pi n^*\right)\left(1 - \frac{1}{6}\pi n^*\right)^{-1}, \tag{53}$$

$$\chi_0 = \frac{1}{1-\xi} + \frac{\pi}{4}\frac{n^*}{(1-\xi)^2}\left(\frac{\sigma_0^*}{\bar{\sigma}^*}\right) + \frac{\pi^2}{72}\frac{n^{*2}}{(1-\xi)^3}\left(\frac{\sigma_0^*}{\bar{\sigma}^*}\right)^2, \tag{54}$$

where $\xi = (\pi n/6)\,\sigma^3 = (\pi/6)\,n^*$ is the volume packing fraction. The influence of density on the Einstein ratio is not as strong as that found for the mass ratio, although it seen to be significant.

5. DISCUSSION

The analysis here has been based on a formally exact description of transport (diffusion and mobility) in the homogeneous cooling state. Green–Kubo expressions for the transport coefficients were obtained by extending familiar linear response methods for fluids with elastic collisions to granular fluids. There are some complications due to the time dependence of the reference state, necessitating a change of time scale. In the resulting dimensionless representation the modified Liouville equation supports a stationary solution, the homogeneous cooling state (HCS), and application of linear response is then more straightforward. However, in addition to the nonlinear change in time scale there appears explicitly in the modified Liouville equation a new fundamental frequency, the cooling rate. The time dependence of the response functions and their relationship to transport coefficients is therefore more complex. The approximate cumulant expansion exposes this complexity in two ways. First, the approximate decay is exponential in the new time scale s, implying algebraic decay in real time. Second, the characteristic frequency for this exponential decay is the difference between an impurity–fluid collision frequency and the cooling rate, $\omega_D^* = \nu^* - \frac{1}{2}\zeta^*$. In the HCS, $\zeta^* = \zeta_0^*$, where ζ_0^* is the corresponding cooling rate for the impurity particle. This equality determines the temperature ratio T_0/T and it is possible to show that $\omega_D^* > 0$ for all values of the density and restitution coefficients. There is a peculiar "phase transition" in the limit of large impurity/fluid mass ratio for which the diffusion coefficient is normal in one phase and grows without bound in the other.[17]

The deviation of the Einstein ratio from unity has three distinct origins. These can be isolated explicitly in the approximate form (44). The deviation of the homogeneous cooling state from the Gibbs state is responsible for the coefficients c_0 and c_1 being non zero. This is a relatively small effect quantitatively speaking. The effect of cooling on the time integral of the response functions for the mobility coefficient occurs through the shift of ω_μ^* to $\omega_\mu^* + \zeta^*$ in the numerator. This feature is described phenomenologically in Section 2. Finally, the effect of different temperatures for the impurity and fluid particles is expressed by the prefactor in Eq. (44) and the factors of θ in ω_D^* and ω_μ^*. Each effect is a different reflection of dissipation, of course, but the Einstein coefficient provides an interesting explicit illustration of each effect separately in the analysis.

In summary, there is growing theoretical support for the validity of hydrodynamic transport processes in granular fluids. However, in detail some care is warranted in translating properties of normal fluids to those with inelastic collisions. As demonstrated here, the familiar relationships between diffusion, mobility, and friction constants no longer hold. The modifications obtained above may be of some importance in choosing parameters in sedimentation problems, for example.

APPENDIX A. ADJOINT LIOUVILLE OPERATORS

The average of some phase function $A(\Gamma)$ at time t is given by

$$\langle A; t\rangle = \int d\Gamma\ \rho(\Gamma, t)\ A(\Gamma) = \int d\Gamma [e^{-\bar{L}t}\rho(\Gamma)]\ A(\Gamma), \tag{A1}$$

where the Liouville operator $\bar{L}$ is defined following Eq. (9). An adjoint Liouville operator L is defined by

$$\langle A; t\rangle = \int d\Gamma [e^{-\bar{L}t}\rho(\Gamma)]\ A(\Gamma) = \int d\Gamma [\rho(\Gamma)\ e^{Lt}A(\Gamma)], \tag{A2}$$

and is found to be

$$L = \sum_{i=0}^{N} \mathbf{v}_i \cdot \nabla_i + \sum_{i=1}^{N} T(i, 0) + \frac{1}{2}\sum_{i=1}^{N}\sum_{j\neq i}^{N} T(i, j), \tag{A3}$$

$$T(i, j) = -\sigma^2 \int d\Omega\ \Theta(-\mathbf{g}_{ij}\cdot\hat{\boldsymbol{\sigma}})(\mathbf{g}_{ij}\cdot\hat{\boldsymbol{\sigma}})\ \delta(\mathbf{r}_{ij} - \hat{\boldsymbol{\sigma}})(b_{ij} - 1), \tag{A4}$$

$$T(i, 0) = -\bar{\sigma}^2 \int d\Omega\ \Theta(-\mathbf{g}_{i0}\cdot\hat{\boldsymbol{\sigma}})(\mathbf{g}_{i0}\cdot\hat{\boldsymbol{\sigma}})\ \delta(\mathbf{r}_{i0} - \bar{\boldsymbol{\sigma}})(b_{i0} - 1), \tag{A5}$$

where

$$b_{ij}\mathbf{g}_{ij} = \mathbf{g}_{ij} - (1+\alpha)\ \hat{\boldsymbol{\sigma}}(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{ij}), \qquad b_{i0}\mathbf{g}_{i0} = \mathbf{g}_{i0} - (1+\alpha_0)(\hat{\boldsymbol{\sigma}}\cdot\mathbf{g}_{i0})\ \hat{\boldsymbol{\sigma}}. \tag{A6}$$

In a similar way an adjoint operator is associated with $\mathscr{L}$ in Eq. (20) by

$$\langle A; t\rangle = \int d\Gamma [e^{-\mathscr{L}t}\rho(\Gamma)]\ A(\Gamma) = \int d\Gamma \rho(\Gamma)[e^{\mathscr{L}t}A(\Gamma)] \tag{A7}$$

with

$$\mathscr{L} \equiv L + \tfrac{1}{2}\zeta(t) \sum_{i=0}^{N} \mathbf{v}_i \cdot \nabla_{v_i} \tag{A8}$$

Finally, its dimensionless form is

$$\mathscr{L}^* \equiv L^* + \tfrac{1}{2}\zeta^* \sum_{i=0}^{N} \mathbf{v}_i^* \cdot \nabla_{v_i^*}. \tag{A9}$$

APPENDIX B. EVALUATION OF ω_D^*, ω_μ^*, AND ζ^*

The diffusion and mobility coefficients are determined from the dimensionless frequencies ω_D^*, ω_μ^*, and ζ^* given by Eqs. (38), (39), and the dimensionless form of Eq. (18). These can be simplified by using the results of Appendix A. In the case of ω_D^* one gets

$$\begin{aligned}
\omega_D^* &= -\frac{\langle (\mathscr{L}^* \mathbf{v}_0^*) \cdot \mathbf{v}_0^* \rangle_{\text{hcs}}}{\langle v_0^{*2} \rangle_{\text{hcs}}} \\
&= -\frac{1}{2}\zeta^* - \frac{N}{\langle v_0^{*2} \rangle_{\text{hcs}}} \langle (T^*(1,0)\, \mathbf{v}_0^*) \cdot \mathbf{v}_0^* \rangle_{\text{hcs}} \\
&= -\frac{1}{2}\zeta^* + \frac{2}{3}\theta \bar{\sigma}^{*2} \int d\mathbf{v}_1^* \int d\mathbf{v}_0^*\, \mathbf{v}_0^* \cdot \int d\Omega\, \Theta(-\mathbf{g}_{10}^* \cdot \hat{\boldsymbol{\sigma}}) \\
&\quad \times f^{*(2)}(\mathbf{v}_1^*, \mathbf{v}_0^*, \mathbf{r}_{10} = \bar{\boldsymbol{\sigma}})(\mathbf{g}_{10}^* \cdot \hat{\boldsymbol{\sigma}})(b_{10} - 1)\, \mathbf{v}_0^* \\
&= -\frac{1}{2}\zeta^* + \frac{2}{3}\frac{m\theta\bar{\sigma}^{*2}}{(m + m_0)}(1 + \alpha_0) \int d\mathbf{v}_1^* \int d\mathbf{v}_0^*\, \mathbf{v}_0^* \cdot \int d\Omega\, \Theta(-\mathbf{g}_{10}^* \cdot \hat{\boldsymbol{\sigma}}) \\
&\quad \times f^{*(2)}(\mathbf{v}_1^*, \mathbf{v}_0^*, \mathbf{r}_{10} = \bar{\boldsymbol{\sigma}})(\mathbf{g}_{10}^* \cdot \hat{\boldsymbol{\sigma}})^2\, \hat{\boldsymbol{\sigma}},
\end{aligned} \tag{B1}$$

where $f^{*(2)}(\mathbf{v}_1^*, \mathbf{v}_0^*, \mathbf{r}_{10} = \bar{\boldsymbol{\sigma}})$ is the reduced two particle distribution function for the HCS evaluated at contact. Use has been made of the second equality in Eq. (43), which defines the temperature of the impurity particle $T_0(t)$ in the HCS. This temperature differs from that of the fluid $T(t)$. However, the cooling rates are the same so that the temperature ratio $T_0(t)/T(t)$ is time independent. Similarly,

$$\omega_\mu^* = \frac{1}{3}\left\langle (\mathscr{L}^*\mathbf{v}_0^*)\cdot\frac{\partial \ln \rho_{\rm hcs}^*}{\partial \mathbf{v}_0^*}\right\rangle_{\rm hcs}$$

$$= -\frac{1}{2}\zeta^* + \frac{N}{3}\left\langle (T^*(1,0)\,\mathbf{v}_0^*)\cdot\frac{\partial \ln \rho_{\rm hcs}^*}{\partial \mathbf{v}_0^*}\right\rangle_{\rm hcs}$$

$$= -\frac{1}{2}\zeta^* - \frac{\bar{\sigma}^{*2}}{3}\int d\mathbf{v}_1^*\int d\mathbf{v}_0^*\int d\Omega\,\Theta(-\mathbf{g}_{10}^*\cdot\hat{\boldsymbol{\sigma}})\left[\frac{\partial}{\partial \mathbf{v}_0^*} f^{*(2)}(\mathbf{v}_1^*, \mathbf{v}_0^*, \mathbf{r}_{10}=\bar{\boldsymbol{\sigma}})\right]$$

$$\times(\mathbf{g}_{10}^*\cdot\hat{\boldsymbol{\sigma}})(b_{10}-1)\,\mathbf{v}_0^*$$

$$= -\frac{1}{2}\zeta^* - \frac{1}{3}\frac{m\bar{\sigma}^{*2}}{(m+m_0)}(1+\alpha_0)\int d\mathbf{v}_1^*\int d\mathbf{v}_0^*\int d\Omega\,\Theta(-\mathbf{g}_{10}^*\cdot\hat{\boldsymbol{\sigma}})(\mathbf{g}_{10}^*\cdot\hat{\boldsymbol{\sigma}})^2$$

$$\times\hat{\boldsymbol{\sigma}}\cdot\left[\frac{\partial}{\partial \mathbf{v}_0^*} f^{*(2)}(\mathbf{v}_1^*, \mathbf{v}_0^*, \mathbf{r}_{10}=\bar{\boldsymbol{\sigma}})\right]. \tag{B2}$$

Finally, the cooling rate is

$$\zeta^* = (1-\alpha^2)\,\tfrac{1}{6}\,\sigma^{*4}\int d\mathbf{v}_1^*\int d\mathbf{v}_2^*\int d\Omega\,\Theta(\mathbf{g}_{ij}^*\cdot\hat{\boldsymbol{\sigma}})(\mathbf{g}_{ij}^*\cdot\hat{\boldsymbol{\sigma}})^3\, f^{*(2)}(\mathbf{v}_1^*, \mathbf{v}_2^*, \mathbf{r}_{12}=\boldsymbol{\sigma}). \tag{B3}$$

Further simplifications are possible if velocity correlations are neglected and the first Sonine approximation for the one particle distribution functions is used. In this case, one has

$$f^{*(2)}(\mathbf{v}_1^*, \mathbf{v}_0^*, \mathbf{r}_{10}=\bar{\boldsymbol{\sigma}}) \to \chi_0\sigma^{*-2} f^{*(1)}(\mathbf{v}_1^*)\, f_0^{*(1)}(\mathbf{v}_0^*), \tag{B4}$$

$$f^{*(2)}(\mathbf{v}_1^*, \mathbf{v}_2^*, \mathbf{r}_{12}=\boldsymbol{\sigma}) \to \chi\sigma^{*-4} f^{*(1)}(\mathbf{v}_1^*)\, f^{*(1)}(\mathbf{v}_2^*), \tag{B5}$$

$$f^{*(1)}(\mathbf{v}_1^*) = \left(\frac{1}{\pi}\right)^{3/2} e^{-v_1^{*2}}\left[1+\frac{c_1}{4}\left(v_1^{*4}-5v_1^{*2}+\frac{15}{4}\right)\right], \tag{B6}$$

$$f_0^{*(1)}(\mathbf{v}_0^*) = \left(\frac{\theta}{\pi}\right)^{3/2} e^{-\theta v_0^{*2}}\left[1+\frac{c_0}{4}\left(\theta^2 v_0^{*4}-5\theta v_0^{*2}+\frac{15}{4}\right)\right]. \tag{B7}$$

Here, χ_0 is the pair correlation function for the fluid–impurity reduced distribution function evaluated at contact, χ is the corresponding fluid–fluid distribution, and $\theta = Tm_0/T_0m$. The coefficient c_1 is[16]

$$c_1(\alpha) = \frac{32(1-\alpha)(1-2\alpha^2)}{81-17\alpha+30\alpha^2(1-\alpha)}. \tag{B8}$$

The coefficient c_0 is more complex and has a dependence on the mass ratio, size ratio, and both restitution coefficients. Its calculation is described in ref. 11 (If the impurity particle is mechanically the same as the fluid particles then $c_0 \to c_1$). Neglecting velocity correlations, Eqs. (B1)–(B3) become

$$\omega_D^* = -\frac{1}{2}\zeta^* + \frac{2}{3}\theta\frac{\bar{\sigma}^{*2}}{\sigma^{*2}}\frac{m}{(m+m_0)}(1+\alpha_0)\,\chi_0 \int d\mathbf{v}_1^* \int d\mathbf{v}_0^*\, f^{*(1)}(\mathbf{v}_1^*)\, f_0^{*(1)}(\mathbf{v}_0^*)$$

$$\times \mathbf{v}_0^* \cdot \int d\Omega\, \Theta(-\mathbf{g}_{10}^* \cdot \hat{\boldsymbol{\sigma}})(\mathbf{g}_{10}^* \cdot \hat{\boldsymbol{\sigma}})^2\, \hat{\boldsymbol{\sigma}}$$

$$= -\frac{1}{2}\zeta^* + \frac{\pi}{3}\theta\frac{\bar{\sigma}^{*2}}{\sigma^{*2}}\frac{m}{(m+m_0)}(1+\alpha_0)\,\chi_0 I_D, \tag{B9}$$

$$\omega_\mu^* = -\frac{1}{2}\zeta^* - \frac{1}{3}\frac{\bar{\sigma}^{*2}}{\sigma^{*2}}\frac{m}{(m+m_0)}(1+\alpha_0)\,\chi_0 \int d\mathbf{v}_1^* \int d\mathbf{v}_0^*\, f^{*(1)}(\mathbf{v}_1^*)$$

$$\times \left[\frac{\partial f_0^{*(1)}(\mathbf{v}_0^*)}{\partial \mathbf{v}_0^*}\right] \cdot \int d\Omega\, \Theta(-\mathbf{g}_{10}^* \cdot \hat{\boldsymbol{\sigma}})(\mathbf{g}_{10}^* \cdot \hat{\boldsymbol{\sigma}})^2\, \hat{\boldsymbol{\sigma}}$$

$$= -\frac{1}{2}\zeta^* + \frac{2\pi}{3}\frac{\bar{\sigma}^{*2}}{\sigma^{*2}}\frac{m}{(m+m_0)}(1+\alpha_0)\,\chi_0 I_\mu, \tag{B10}$$

$$\zeta^* = (1-\alpha^2)\frac{1}{6}\chi \int d\mathbf{v}_1^* \int d\mathbf{v}_2^*\, f^{*(1)}(\mathbf{v}_1^*)\, f^{*(1)}(\mathbf{v}_2^*) \int d\Omega\, \Theta(\mathbf{g}_{12}^* \cdot \hat{\boldsymbol{\sigma}})(\mathbf{g}_{12}^* \cdot \hat{\boldsymbol{\sigma}})^3$$

$$= (1-\alpha^2)\,\chi\frac{\pi}{12} I_\zeta. \tag{B11}$$

The integrals I_D, I_μ, and I_ζ are defined by

$$I_D = -\int d\mathbf{v}_1^* \int d\mathbf{v}_0^*\, f^{*(1)}(\mathbf{v}_1^*)\, f_0^{*(1)}(\mathbf{v}_0^*)(\mathbf{v}_0^* \cdot \mathbf{g}_{10}^*)\, g_{10}^*, \tag{B12}$$

$$I_\mu = \int d\mathbf{v}_1^* \int d\mathbf{v}_0^*\, f^{*(1)}(\mathbf{v}_1^*)\, f_0^{*(1)}(\mathbf{v}_0^*)\, g_{10}^*, \tag{B13}$$

$$I_\zeta = \int d\mathbf{v}_1^* \int d\mathbf{v}_2^*\, f^{*(1)}(\mathbf{v}_1^*)\, f^{*(1)}(\mathbf{v}_2^*)\, g_{12}^{*3}. \tag{B14}$$

Consider first the evaluation of I_ζ. Introduce the approximate distribution (B6) and retain terms up through linear in c_1, consistent with the Sonine polynomial expansion

$$I_\zeta = \pi^{-3} \int d\mathbf{v}_1^* \int d\mathbf{v}_2^* \, e^{-(v_1^{*2}+v_2^{*2})} g_{12}^{*3} \left[1 + \frac{c_1}{4} \left(v_1^{*4} - 5v_1^{*2} + \frac{15}{4} + v_2^{*4} - 5v_2^{*2} + \frac{15}{4} \right) \right]. \tag{B15}$$

The integration is straightforward with a change of variables to relative and center of mass $\mathbf{v}_1^* = (\mathbf{y} + \frac{1}{2}\mathbf{x})$ and $\mathbf{v}_2^* = (\mathbf{y} - \frac{1}{2}\mathbf{x})$ with the result

$$I_\zeta = \frac{16}{\sqrt{2\pi}} \left(1 + \frac{3}{32} c_1 \right). \tag{B16}$$

The cooling rate becomes

$$\zeta^* = (1-\alpha^2) \, \chi \, \frac{4}{3} \sqrt{\frac{\pi}{2}} \left(1 + \frac{3}{32} c_1 \right). \tag{B17}$$

The integrals I_D and I_μ are somewhat more complicated. Introducing the distribution functions and retaining terms up through linear in c_0 and c_1

$$\begin{aligned} I_D &= -\left(\frac{\theta\theta_1}{\pi^2} \right)^{3/2} \int d\mathbf{v}_1^* \int d\mathbf{v}_0^* \, e^{-(\theta_1 v_1^{*2} + \theta v_0^{*2})} g_{10}^* (\mathbf{v}_0^* \cdot \mathbf{g}_{10}^*) \\ &\quad \times \left[1 + \frac{c_0}{4} \left(\theta^2 v_0^{*4} - 5\theta v_0^{*2} + \frac{15}{4} \right) + \frac{c_1}{4} \left(\theta_1^2 v_1^{*4} - 5\theta_1 v_1^{*2} + \frac{15}{4} \right) \right] \\ &= \left(\frac{\theta\theta_1}{\pi^2} \right)^{3/2} \left\{ 1 + \frac{c_0}{4} \left(\theta^2 \frac{d^2}{d\theta^2} + 5\theta \frac{d}{d\theta} + \frac{15}{4} \right) \right. \\ &\quad \left. + \frac{c_1}{4} \left(\theta_1^2 \frac{d^2}{d\theta_1^2} + 5\theta_1 \frac{d}{d\theta_1} + \frac{15}{4} \right) \right\} J_D(\theta, \theta_1) \end{aligned} \tag{B18}$$

where

$$J_D(\theta, \theta_1) = -\int d\mathbf{v}_1^* \, d\mathbf{v}_0^* \, e^{-(\theta_1 v_1^{*2} + \theta v_0^{*2})} g_{10}^* (\mathbf{v}_0^* \cdot \mathbf{g}_{10}^*) \tag{B19}$$

and the parameter $\theta_1 \to 1$ after the differentiations. Similarly,

$$I_\mu = \left(\frac{\theta\theta_1}{\pi^2}\right)^{3/2} \left\{1+\frac{c_0}{4}\left(\theta^2\frac{d^2}{d\theta^2}+5\theta\frac{d}{d\theta}+\frac{15}{4}\right) + \frac{c_1}{4}\left(\theta_1^2\frac{d^2}{d\theta_1^2}+5\theta_1\frac{d}{d\theta_1}+\frac{15}{4}\right)\right\} J_\mu(\theta,\theta_1), \tag{B20}$$

$$J_\mu(\theta,\theta_1) = \int d\mathbf{v}_1^* \int d\mathbf{v}_0^*\, e^{-(\theta_1 v_1^{*2}+\theta v_0^{*2})} g_{10}^*. \tag{B21}$$

The integrals $J_D(\theta,\theta_1)$ and $J_\mu(\theta,\theta_1)$ can be performed by the change of variables

$$\mathbf{x} = \mathbf{v}_1^* - \mathbf{v}_0^*, \qquad \mathbf{y} = \theta_1\mathbf{v}_1^* + \theta\mathbf{v}_0^*, \tag{B22}$$

with the Jacobian $(\theta_1+\theta)^{-3}$. The integrals become

$$J_D(\theta,\theta_1) = \theta_1(\theta_1+\theta)^{-4}\int d\mathbf{x}\, x^3 e^{-ax^2}\int d\mathbf{y}\, e^{-by^2}, \tag{B23}$$

$$J_\mu(\theta,\theta_1) = (\theta_1+\theta)^{-3}\int d\mathbf{x}\, x e^{-ax^2}\int d\mathbf{y}\, e^{-by^2}, \tag{B24}$$

where

$$a = (\theta_1+\theta)^{-1}\theta\theta_1, \qquad b = (\theta_1+\theta)^{-1}. \tag{B25}$$

The integrals are easily performed, with the results

$$J_D(\theta,\theta_1) = 4\pi^{5/2}\frac{(\theta_1+\theta)^{1/2}}{\theta^3\theta_1^2}, \qquad J_\mu(\theta,\theta_1) = 2\pi^{5/2}\frac{(\theta_1+\theta)^{1/2}}{\theta^2\theta_1^2} \tag{B26}$$

The integrals I_D and I_μ follow directly from (B18) and (B20)

$$I_D = 4\theta^{-3/2}\pi^{-1/2}(1+\theta)^{1/2}\left[1+\frac{c_0}{16}\frac{4\theta+3}{(1+\theta)^2}-\frac{c_1}{16}\frac{\theta^2}{(1+\theta)^2}\right], \tag{B27}$$

$$I_\mu = 2\pi^{-1/2}\theta^{-1/2}(1+\theta)^{1/2}\left[1-\frac{c_0}{16}\frac{1}{(1+\theta)^2}-\frac{c_1}{16}\frac{\theta^2}{(1+\theta)^2}\right]. \tag{B28}$$

Finally, use of these results in Eqs. (B9) and (B10) gives the desired results

$$\omega_D^* = -\frac{1}{2}\zeta^* + \nu^* \left[1 + \frac{c_0}{16}\frac{4\theta+3}{(1+\theta)^2} - \frac{c_1}{16}\frac{\theta^2}{(1+\theta)^2} \right], \tag{B29}$$

$$\omega_\mu^* = -\frac{1}{2}\zeta^* + \nu^* \left[1 - \frac{c_0}{16}\frac{1}{(1+\theta)^2} - \frac{c_1}{16}\frac{\theta^2}{(1+\theta)^2} \right], \tag{B30}$$

$$\nu^* = \frac{4\sqrt{\pi}}{3}\frac{m}{(m+m_0)}\chi_0 \frac{\bar{\sigma}^2}{\sigma^2}(1+\alpha_0)\,\theta^{-1/2}(1+\theta)^{1/2}. \tag{B31}$$

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