

**Diffusion in a granular fluid. I. Theory**

James W. Dufty

*Department of Physics, University of Florida, Gainesville, Florida 32611*

J. Javier Brey

*Física Teórica, Universidad de Sevilla, Apartado de Correos 1065, E-41080 Sevilla, Spain*

James Lutsko

*Center for Nonlinear Phenomena and Complex Systems, Université Libre de Bruxelles, Campus Plaine, CP 231, 1050-Bruxelles, Belgium*

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Many important properties of granular fluids can be represented by a system of hard spheres with inelastic collisions. Traditional methods of nonequilibrium statistical mechanics are effective for analysis and description of the inelastic case as well. This is illustrated here for diffusion of an impurity particle in a fluid undergoing homogeneous cooling. An appropriate scaling of the Liouville equation is described such that the homogeneous cooling ensemble and associated time correlation functions map to those of a stationary state. In this form the familiar methods of linear response can be applied, leading to Green-Kubo and Einstein representations of diffusion in terms of the velocity and mean-square displacement correlation functions. These correlation functions are evaluated approximately using a cumulant expansion and from kinetic theory, providing the diffusion coefficient as a function of the density and the restitution coefficients. Comparisons with results from molecular-dynamics simulation are given in the following companion paper.

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**I. INTRODUCTION**

It has long been recognized that rapid flow granular media have properties similar to those of ordinary fluids [1,2]. Attempts to sharpen this relationship have used idealized systems of hard spheres with inelastic collisions. Remarkably, the single feature of inelasticity allows reproduction of many qualitative features observed in real granular fluids. This observation is based on the derivation of hydrodynamic equations from kinetic theory [3–7], direct Monte Carlo simulation of kinetic equations [8,9], molecular-dynamics simulation of dense fluids [10], and controlled experiments on inelastic hard objects such as spherical beads [11–16]. Consequently, there is a growing opinion that the traditional methods of nonequilibrium statistical mechanics applied to such model systems provide the means to understand granular media at the most fundamental microscopic level. The objective here is to give further support for this opinion by a detailed application and test of linear response methods applied to diffusion of an impurity particle in a fluid of inelastic hard spheres. Some adaptation is required since the reference states are inherently nonequilibrium, but the central ideas of linear response for normal fluids are retained. A preliminary report of some of these results presented here has been given in [17].

Diffusion is the prototype transport process and the associated diffusion equation is the prototype hydrodynamic description for macroscopic dynamics. For normal fluids, diffusion in a system of hard elastic spheres also has been the testing ground for many-body methods in nonequilibrium statistical mechanics (density dependence, correlated many-body collisions, mode coupling, percolation, glass transition). The benchmarks have been set by accurate molecular-dynamics simulation for normal fluids [18] and fluidlike

lattice-gas cellular automata [19]. In this way, the conditions for macroscopic diffusion and the accuracy of methods for predicting the diffusion coefficient are known over a wide range of densities. There is evidence based on kinetic theory and molecular-dynamics simulation that similar studies are relevant for the system of inelastic hard spheres [20–22]. On the other hand, there are significant differences to confront. An isolated fluid of inelastic hard spheres does not support a Gibbs state or any other stationary state, since the collisions lead to a continual loss of energy or “cooling.” Instead, the analog of the equilibrium state is a “homogeneous cooling state” (HCS) whose time dependence is entirely given via the mean-square kinetic energy of the particles [23,24,8,25]. As shown below, the scaling property associated with this state allows a change of variables in terms of which a stationary, but non-Gibbs, state results.

In the next section, the Liouville dynamics for inelastic hard spheres is reviewed [17,24,22]. The corresponding nonequilibrium statistical mechanics is given in terms of the Liouville equation for the ensemble, and also in terms of the Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) hierarchy for the associated reduced distribution functions. Next, it is shown that the Liouville equation supports a scaling solution describing the HCS. A time-dependent temperature is defined in terms of the mean-square velocity, in the same way as for elastic collisions, hence the terminology “homogeneous cooling state.” A transformation to dimensionless variables allows a representation of expectation values and time correlation functions in terms of stationary state averages, just as for the Gibbs state.

The probability density for the position of a tagged or impurity particle as a function of time is considered in Sec. III. Linear-response methods (now inherently nonequilibrium linear response) are applied to obtain a diffusionlike equation for long wavelengths, with a time-dependent diffusion coef-

ficient. The analysis parallels closely that for fluids with elastic collisions, except that the dimensionless time (the collision number) required to accommodate the cooling is logarithmically stretched compared to real time. Consequently, the mean-square displacement approaches linearity in the collision number rather than real time. The diffusion coefficient is expressed as a time integral of the velocity autocorrelation function which becomes a Green-Kubo expression for long times. This limit also establishes the time scale for the validity of the diffusion equation, and represents a clear example of macroscopic hydrodynamics for a granular system.

An approximate evaluation of the velocity correlation function of the tagged particle is carried out in Sec. IV by two different methods. The first is based on a leading cumulant expansion, while the second is an evaluation by means of kinetic theory. The two results are quite similar, and their relationship is clearly identified. Also, it is confirmed in Appendix B that the evaluation of the Green-Kubo expression by kinetic theory agrees with that obtained by the Chapman-Enskog solution to the Enskog kinetic equation for the distribution function. Moreover, for elastic collisions these approximations are known to give an accurate description of the diffusion coefficient over a wide range of densities. This accuracy cannot be assumed *a priori* to apply for inelastic collisions, since the stationary state is nonequilibrium. The diffusion coefficient is given as a function of the density, restitution coefficient, and ratio of temperatures for the fluid and impurity particles. This latter parameter is a peculiarity of granular fluids for mixtures, where the lack of detailed balance leads to a HCS with all species having the same cooling rate but different temperatures [26]. The consequences for impurity diffusion and mobility have been discussed elsewhere recently [27,28]. The last section contains a summary and discussion of the main results. The theoretical developments presented here are tested and extended in a companion paper by comparison with molecular dynamics simulation results over a wide range of densities and inelasticities for the particular case of self diffusion [29].

## II. STATISTICAL MECHANICS AND HOMOGENEOUS COOLING STATE

The system considered is composed of a fluid of  $N$  identical hard disks or spheres (mass  $m$ , diameter  $\sigma$ , and fluid-fluid coefficient of normal restitution  $\alpha$ ), and an additional impurity particle (mass  $m_0$ , diameter  $\sigma_0$ , and fluid-impurity coefficient of normal restitution  $\alpha_0$ ). The position and velocity coordinates of the fluid particles will be denoted by  $\{\mathbf{q}_i, \mathbf{v}_i; i=1, \dots, N\}$ , while those for the impurity particle are  $\mathbf{q}_0, \mathbf{v}_0$ . The dynamics consists of free streaming until a given pair of fluid particles,  $i, j$ , or a fluid and impurity pair,  $0, i$ , is in contact. At that point, the relative velocity of the pair changes instantaneously according to the inelastic collision rules

$$\begin{aligned}\tilde{\mathbf{g}}_{ij} &= \mathbf{g}_{ij} - (1 + \alpha)(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{ij})\hat{\boldsymbol{\sigma}}, \\ \tilde{\mathbf{g}}_{0i} &= \mathbf{g}_{0i} - (1 + \alpha_0)(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{0i})\hat{\boldsymbol{\sigma}}.\end{aligned}\quad (1)$$

Here,  $\mathbf{g}_{ij} = \mathbf{v}_i - \mathbf{v}_j$  and  $\mathbf{g}_{0i} = \mathbf{v}_0 - \mathbf{v}_i$  are the relative velocities, and  $\hat{\boldsymbol{\sigma}}$  is a unit vector directed from the center of particle  $j$  to

the center of particle  $i$  ( $i$  to  $0$ , respectively) through the point of contact. The coefficients of restitution have values in the range  $0 < \alpha, \alpha_0 \leq 1$ , measuring the degree of inelasticity. The special case of elastic particles is given by  $\alpha = \alpha_0 = 1$ . The center-of-mass velocity is unchanged so that the total mass and momentum of the pairs are conserved in such collisions. However, there is an energy loss for each fluid-fluid particle collision

$$\tilde{E}_{ij} - E_{ij} = -\frac{m}{4}(1 - \alpha^2)(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{ij})^2 \quad (2)$$

and for each fluid-impurity particle collision

$$\tilde{E}_{0i} - E_{0i} = -\frac{\mu}{2}(1 - \alpha_0^2)(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{0i})^2, \quad (3)$$

where  $\mu = m_0 m / (m_0 + m)$ . The state of the system at time  $t$  is completely characterized by the positions and velocities of all particles at that time and is represented by a point  $\Gamma_t \equiv \{\mathbf{q}_0(t), \dots, \mathbf{q}_N(t), \mathbf{v}_0(t), \dots, \mathbf{v}_N(t)\}$  in the associated  $2d(N+1)$ -dimensional phase space, where  $d=2$  for hard disks and  $d=3$  for hard spheres. The sequence of free streaming and binary collisions determines uniquely the positions and velocities of the hard particles at time  $t$  for given initial conditions at  $t' < t$ . A more complete notation expressing this dependence on initial conditions is  $\Gamma_t(\Gamma_{t'})$ . Thus, just as in the case of elastic collisions, the microdynamics for this system corresponds to a deterministic trajectory in phase space.

Observables of interest are represented by the same phase functions as for elastic collisions,  $A(\Gamma_t)$ , and their average for given statistical initial data at  $t=0$  is defined by

$$\langle A(t); 0 \rangle \equiv \int d\Gamma \rho(\Gamma) A[\Gamma_t(\Gamma)], \quad (4)$$

where  $\rho(\Gamma)$  is the probability density or ensemble for the initial state, normalized to unity. An equivalent representation of this average is obtained by changing variables to integrate over  $\Gamma_t$  rather than over  $\Gamma$ . This change of variables is possible since trajectories in phase space do not cross, and  $\Gamma$  can be expressed in terms of  $\Gamma_t$  denoted by  $\Gamma_t^{-1}(\Gamma_t)$ . This allows the time dependence in Eq. (4) to be expressed in terms of the probability density

$$\langle A(t); 0 \rangle = \int d\Gamma \rho(\Gamma, t) A(\Gamma) \equiv \langle A; t \rangle, \quad (5)$$

with the probability density at time  $t$  given by

$$\rho(\Gamma, t) \equiv J[\Gamma_t^{-1}(\Gamma), \Gamma] \rho[\Gamma_t^{-1}(\Gamma)], \quad (6)$$

$J(\Gamma, \Gamma_t)$  being the Jacobian of the transformation.

### A. Liouville dynamics

For practical purposes it is useful to identify the generators  $L$  and  $\bar{L}$  for the two above representations, defined by

$$\langle A(t); 0 \rangle = \int d\Gamma \rho(\Gamma) e^{tL} A(\Gamma) = \int d\Gamma [e^{-t\bar{L}} \rho(\Gamma)] A(\Gamma). \quad (7)$$

The last equality is consistent with the adjoint relationship implied by Eqs. (4) and (5). These are not the usual generators of Hamilton's equations for continuous forces, but are somewhat more complex due to the singular nature of hard particles. Such generators have been discussed in detail for the case of elastic collisions and the analysis extends quite naturally to the inelastic case as well [24,30,17,22], with the results

$$\begin{aligned} L &= L_f + \mathbf{v}_0 \cdot \nabla_0 + \sum_{i=1}^N T(0,i), \\ \bar{L} &= \bar{L}_f + \mathbf{v}_0 \cdot \nabla_0 - \sum_{i=1}^N \bar{T}(0,i). \end{aligned} \quad (8)$$

Here,  $L_f$  and  $\bar{L}_f$  are the generators for the fluid particles alone

$$\begin{aligned} L_f &= \sum_{i=1}^N \mathbf{v}_i \cdot \nabla_i + \frac{1}{2} \sum_{i=1}^N \sum_{j \neq i}^N T(i,j), \\ \bar{L}_f &= \sum_{i=1}^N \mathbf{v}_i \cdot \nabla_i - \frac{1}{2} \sum_{i=1}^N \sum_{j \neq i}^N \bar{T}(i,j). \end{aligned} \quad (9)$$

The terms involving spatial gradients generate free streaming while the others describe velocity changes. The binary collision operators  $T(i,j)$  and  $\bar{T}(i,j)$  for particles  $i$  and  $j$  are

$$T(i,j) = \sigma^{d-1} \int d\Omega \Theta(-\mathbf{g}_{ij} \cdot \hat{\boldsymbol{\sigma}}) |\mathbf{g}_{ij} \cdot \hat{\boldsymbol{\sigma}}| \delta(\mathbf{q}_{ij} - \boldsymbol{\sigma}) (b_{ij} - 1), \quad (10)$$

$$\begin{aligned} \bar{T}(i,j) &= \sigma^{d-1} \int d\Omega \Theta(\mathbf{g}_{ij} \cdot \hat{\boldsymbol{\sigma}}) |\mathbf{g}_{ij} \cdot \hat{\boldsymbol{\sigma}}| \\ &\quad \times [\alpha^{-2} \delta(\mathbf{q}_{ij} - \boldsymbol{\sigma}) b_{ij}^{-1} - \delta(\mathbf{q}_{ij} + \boldsymbol{\sigma})], \end{aligned} \quad (11)$$

where  $d\Omega$  denotes the solid angle element for the unit vector  $\hat{\boldsymbol{\sigma}}$ ,  $\boldsymbol{\sigma} = \sigma \hat{\boldsymbol{\sigma}}$ ,  $\mathbf{q}_{ij}$  is the relative position vector of the two particles,  $\Theta$  is the Heaviside step function, and  $b_{ij}$  is a substitution operator,  $b_{ij} F(\mathbf{g}_{ij}) = F(\tilde{\mathbf{g}}_{ij})$ , which changes the relative velocity  $\mathbf{g}_{ij}$  into its scattered value  $\tilde{\mathbf{g}}_{ij}$ , given by Eq. (1). On the other hand, it does not change the velocity of the center of mass of the two particles. The operator  $b_{ij}^{-1}$  is the inverse of  $b_{ij}$  and characterizes the ‘‘restituting’’ collision. The binary operators for collisions between fluid particles and the impurity are similar to those for collisions among fluid particles,

$$T(0,i) = \bar{\sigma}^{d-1} \int d\Omega \Theta(-\mathbf{g}_{0i} \cdot \hat{\boldsymbol{\sigma}}) |\mathbf{g}_{0i} \cdot \hat{\boldsymbol{\sigma}}| \delta(\mathbf{q}_{0i} - \bar{\boldsymbol{\sigma}}) (b_{0i} - 1), \quad (12)$$

$$\begin{aligned} \bar{T}(0,i) &= \bar{\sigma}^{d-1} \int d\Omega \Theta(\mathbf{g}_{0i} \cdot \hat{\boldsymbol{\sigma}}) |\mathbf{g}_{0i} \cdot \hat{\boldsymbol{\sigma}}| \\ &\quad \times [\alpha_0^{-2} \delta(\mathbf{q}_{0i} - \bar{\boldsymbol{\sigma}}) b_{0i}^{-1} - \delta(\mathbf{q}_{0i} + \bar{\boldsymbol{\sigma}})], \end{aligned} \quad (13)$$

with  $\bar{\sigma} = (\sigma + \sigma_0)/2$  and  $\bar{\boldsymbol{\sigma}} = \bar{\sigma} \hat{\boldsymbol{\sigma}}$ . The explicit forms for  $b_{ij}^{-1}$  and  $b_{0i}^{-1}$  are

$$b_{ij}^{-1} \mathbf{g}_{ij} = \mathbf{g}_{ij} - \frac{1 + \alpha}{\alpha} (\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{ij}) \hat{\boldsymbol{\sigma}}, \quad (14)$$

$$b_{0i}^{-1} \mathbf{g}_{0i} = \mathbf{g}_{0i} - \frac{1 + \alpha_0}{\alpha_0} (\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{0i}) \hat{\boldsymbol{\sigma}}. \quad (15)$$

The dynamics for the phase functions and the equivalent Liouville equation distribution function,  $\rho(\Gamma, t)$ , follow directly from Eq. (7):

$$(\partial_t - L)A(\Gamma, t) = 0, \quad (16)$$

$$(\partial_t + \bar{L})\rho(\Gamma, t) = 0. \quad (17)$$

The BBGKY hierarchy of equations for the reduced distribution functions is obtained by partial integration of the Liouville equation over the position and velocities of  $N-l$  fluid particles,

$$\begin{aligned} [\partial_t + \bar{L}(x_0, \dots, x_l)] f^{(l+1)}(x_0, \dots, x_l; t) \\ = \sum_{i=0}^l \int dx_{l+1} \bar{T}(i, l+1) f^{(l+2)}(x_0, \dots, x_{l+1}; t), \end{aligned} \quad (18)$$

with the reduced distribution functions defined by

$$f^{(l+1)}(x_0, \dots, x_l; t) \equiv N^l \int dx_{l+1} \dots dx_N \rho(\{x_i\}; t), \quad (19)$$

where  $x_i \equiv \{\mathbf{q}_i, \mathbf{v}_i\}$  denotes the position and velocity for particle  $i$  and  $\bar{L}(x_0, \dots, x_l)$  is the Liouville operator for a system of  $l$  fluid particles and the impurity particle. Moreover, the limit of large  $N$  has been considered. The above results in this section provide the basic tools and definitions of non-equilibrium statistical mechanics for granular media [24].

## B. Homogeneous cooling state (HCS)

Stationary solutions to the Liouville Eq. (17) are expected when suitable external forces or boundary conditions are imposed. However, there is no stationary solution for an isolated system, corresponding to the spatially homogeneous Gibbs state, due to the inherent time dependence following from loss of energy in collisions. This can be seen by calculating the rate of change of the mean-square velocity of a fluid particle in an isolated state. For purposes below, the latter is used to define a kinetic temperature according to

$$T(t) \equiv \frac{1}{d} \langle m \mathbf{v}_1^2; t \rangle \equiv \frac{1}{2} m v^2(t). \quad (20)$$

In addition to  $T(t)$ , Eq. (20) defines the associated thermal velocity  $v(t)$  (a factor of Boltzmann's constant usual in elastic systems has been deleted since there is no zeroth law of thermodynamics for granular media; alternatively, to incorporate the elastic limit  $\alpha=1$  the temperature should understood as defined in units such that  $k_B=1$ ). The time dependence of these quantities can be calculated using the Liouville dynamics to get

$$\partial_t T(t) = -\zeta(t)T(t), \quad (21)$$

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

$$\begin{aligned} \zeta(t) &= (1-\alpha^2) \frac{\sigma^{d-1}}{2dNv^2(t)} \int d\mathbf{q}_1 \int d\mathbf{v}_1 \\ &\times \int d\mathbf{v}_2 \int d\Omega \Theta(\mathbf{g}_{12} \cdot \hat{\boldsymbol{\sigma}})(\mathbf{g}_{12} \cdot \hat{\boldsymbol{\sigma}})^3 \\ &\times f^{(2)}(\mathbf{q}_1, \mathbf{v}_1, \mathbf{q}_1 + \boldsymbol{\sigma}, \mathbf{v}_2; t), \end{aligned} \quad (22)$$

$f^{(2)}(\mathbf{q}_1, \mathbf{v}_1, \mathbf{q}_1 + \boldsymbol{\sigma}, \mathbf{v}_2; t)$  being the reduced two-fluid particle distribution function at contact. The latter is in general defined by

$$f^{(2)}(x_1, x_2; t) = \int dx_0 f^{(3)}(x_0, x_1, x_2; t). \quad (23)$$

For a homogeneous system, its spatial dependence occurs only through  $\mathbf{q}_{12}$ . Upon deriving Eq. (22) we have taken into account that the contribution from the impurity particle is negligible in the limit of large  $N$ .

In place of the Gibbs distribution, it is assumed that there is a homogeneous scaling solution  $\rho_{\text{HCS}}(\Gamma, t)$  to the Liouville equation, for which all time dependence occurs through a scaling of the velocity (‘‘cooling’’) with the thermal velocity  $v(t)$  [23,24],

$$\rho_{\text{HCS}}(\Gamma, t) = [\ell v(t)]^{-d(N+1)} \rho_{\text{HCS}}^*(\{\mathbf{q}_{ij}/\ell, \mathbf{v}_i/v(t)\}). \quad (24)$$

The dimensionless distribution function  $\rho_{\text{HCS}}^*$  is invariant under space translations, with the coordinates scaled (arbitrarily) relative to  $\ell \equiv (n\sigma^{d-1})^{-1}$ , which is proportional to the mean free path ( $n$  is the number density of particles). Therefore,  $\rho_{\text{HCS}}(\Gamma, t)$  represents a spatially homogeneous fluid. Substitution of Eq. (24) into the Liouville equation gives

$$\frac{1}{2} \zeta(t) \sum_{i=0}^N \frac{\partial}{\partial \mathbf{v}_i} \cdot (\mathbf{v}_i \rho_{\text{HCS}}) + \bar{L} \rho_{\text{HCS}} = 0. \quad (25)$$

The self-consistent solution to the coupled set of Eqs. (21) and (25) determines the *homogeneous cooling state* (HCS). It is the analog of the Gibbs state for elastic collisions and reduces to it for  $\alpha=1$ . For  $\alpha < 1$ , the exact solution is not known (it is not simply a Gaussian in the velocities as for the Gibbs state) but its existence is supported by results from Monte Carlo [8] and molecular-dynamics simulations [31].

The notation in Eq. (24) does not make explicit the dependence of  $\rho_{\text{HCS}}$  on parameters specific to the impurity particle, such as the mass and size ratios  $m/m_0$  and  $\sigma/\sigma_0$ . Since all velocities have been scaled relative to the thermal velocity determined by the fluid, there is no explicit dependence on the fluid temperature. However, the kinetic temperature of the impurity particle  $T_0$  defined in a manner analogous to Eq. (20) is *not* equal to the temperature of the fluid in the HCS. Consequently, scaling of the impurity velocities to the fluid thermal velocities leads to a dependence on the temperature ratio  $T/T_0$ . This peculiarity is due to a failure of energy equipartition in granular fluids [26], which has been observed in recent experiments [32]. The detailed form of the relationship between  $T_0(t)$  and  $T(t)$  will be discussed later on, but it follows from the condition for the HCS that the cooling rates of the fluid and impurity particles are the same in the HCS. This leads to an explicit expression for  $T/T_0$  as a time-independent function of the mechanical differences between the two types of particles. Through this section, the dependence of  $\rho_{\text{HCS}}$  on time independent parameters of the impurity particle will continue to be suppressed, although it will become important in the subsequent discussion of impurity diffusion.

Some interesting consequences follow from the velocity scaling of the distribution function associated with the HCS. The reduced distribution functions also have this property, so it is easily verified from Eq. (22) that  $\zeta(t) \propto T^{1/2}(t)$ . Then, Eq. (21) can be integrated for the explicit time dependence of  $T(t)$ ,

$$T(t) = T(t') \left[ 1 + \frac{1}{2} \zeta(t')(t-t') \right]^{-2}. \quad (26)$$

The temperature is seen to have an algebraic decay in real time (Haff's law [1]). For the analysis of the HCS, it is more convenient to use the dimensionless time scale

$$s(t, t') \equiv \int_{t'}^t d\tau \nu_c(\tau), \quad (27)$$

where  $\nu_c(t)$  is an average collision frequency given by

$$\nu_c(t) = v(t)/\ell. \quad (28)$$

Thus,  $s(t, t')$  is a measure of the average number of collisions per fluid particle in the interval  $(t', t)$ . The integral in Eq. (27) can be performed using Eq. (26) with the following result:

$$s(t, t') = \frac{2}{\zeta^*} \ln \left[ 1 + \frac{1}{2} \zeta(t')(t-t') \right], \quad (29)$$

where we have introduced the dimensionless cooling rate

$$\zeta^* = \ell \zeta(t)/v(t). \quad (30)$$

It follows from dimensional analysis that  $\zeta^*$  is time independent. The cooling in terms of the dimensionless time  $s$  is exponential,

$$T(t) = T(t') e^{-\zeta^* s(t,t')} \quad (31)$$

and, consequently,

$$v(t) = v(t') e^{-\zeta^* s(t,t')/2}. \quad (32)$$

Since  $\zeta$  is proportional to  $(1 - \alpha^2)$ , there is a crossover from logarithmic to linear relationship between the two time scales for weak inelasticity.

Knowledge of the time dependence of  $v(t)$  also implies that for many average properties. For example, the average value of  $A(\Gamma)$  in the HCS can be written

$$\begin{aligned} \langle A; t \rangle_{\text{HCS}} &= \int d\Gamma \rho_{\text{HCS}}(\Gamma, t) A(\Gamma) \\ &= \int d\Gamma^* \rho_{\text{HCS}}^*(\Gamma^*) A(\{\ell \mathbf{q}_i^*, v(t) \mathbf{v}_i^*\}). \end{aligned} \quad (33)$$

Use has been made of the scaling form (24) and  $\Gamma^* = \{\mathbf{q}_i^*, \mathbf{v}_i^*\} = \{\mathbf{q}_i / \ell, \mathbf{v}_i / v(t)\}$ . This last result suggests that the transformation to dimensionless form may admit a stationary state representation for the HCS. To see that this is the case, define for a general state  $\rho(\Gamma, t)$ ,

$$\rho(\Gamma, t) = [\ell v(t)]^{-d(N+1)} \rho^*(\Gamma^*, s), \quad (34)$$

where we have introduced the same scaling for space, time, and velocity as above. Substitution of this into Eq. (17) gives the dimensionless Liouville equation

$$(\partial_s + \bar{\mathcal{L}}^*) \rho^*(\Gamma^*, s) = 0, \quad (35)$$

with the definitions

$$\bar{\mathcal{L}}^* = \frac{1}{2} \zeta^* [\mathcal{K}^* + d(N+1)] + \bar{\mathcal{L}}^*, \quad (36)$$

$$\bar{\mathcal{L}}^* = \frac{1}{v_c(t)} \bar{\mathcal{L}} = [\bar{\mathcal{L}}]_{\{\mathbf{q}_i = \mathbf{q}_i^*, \mathbf{v}_i = \mathbf{v}_i^*\}}, \quad (37)$$

$$\mathcal{K}^* = \sum_{i=0}^N \mathbf{v}_i^* \cdot \frac{\partial}{\partial \mathbf{v}_i^*}. \quad (38)$$

This transformation of the Liouville equation explicitly accounts for the collisional cooling, and in this form stationary solutions are now possible. In fact, Eq. (25) becomes

$$\bar{\mathcal{L}}^* \rho_{\text{HCS}}^* = 0, \quad (39)$$

so the dimensionless HCS is a stationary solution to Eq. (35).

The average value of  $A(\Gamma)$  for a general state  $\rho(\Gamma, t)$  becomes

$$\begin{aligned} \langle A; t \rangle &= \int d\Gamma \rho(\Gamma, t) A(\Gamma) \\ &= \int d\Gamma^* \rho^*(\Gamma^*, s) A(\{\ell \mathbf{q}_i^*, v(t) \mathbf{v}_i^*\}) \\ &= \int d\Gamma^* [\bar{U}(s, 0) \rho^*(\Gamma^*, 0)] A(\{\ell \mathbf{q}_i^*, v(t) \mathbf{v}_i^*\}) \\ &\equiv \int d\Gamma^* \rho^*(\Gamma^*, 0) U(s, 0) A(\{\ell \mathbf{q}_i^*, v(t) \mathbf{v}_i^*\}), \end{aligned} \quad (40)$$

where  $\bar{U}(s, s')$  and  $U(s, s')$  obey the equations

$$(\partial_s + \bar{\mathcal{L}}^*) \bar{U}(s, s') = 0, \quad (\partial_s - \mathcal{L}^*) U(s, s') = 0, \quad (41)$$

with the initial conditions  $\bar{U}(s', s') = U(s', s') = 1$ . The new generator for the dynamics of the phase functions is

$$\mathcal{L}^* = \frac{1}{2} \zeta^* \mathcal{K}^* + \mathcal{L}^*, \quad \mathcal{L}^* = [\mathcal{L}]_{\{\mathbf{q}_i = \mathbf{q}_i^*, \mathbf{v}_i = \mathbf{v}_i^*\}}. \quad (42)$$

For the special case  $\rho^*(\Gamma^*, 0) = \rho_{\text{HCS}}^*(\Gamma^*)$ , Eq. (40) reduces to Eq. (33).

The stationary representation is the most natural one for both theoretical developments and for computer simulation, as is discussed in the following companion paper [29]. Similarly, the physically relevant time scales are those expressed in terms of the average collision number  $s$  rather than the real time  $t$ . It is appropriate at this point to note that although  $\rho_{\text{HCS}}^*$  is a stationary solution to Eq. (35), there is convincing evidence from both theory and simulation that it is unstable to long wavelength spatial perturbations and spontaneous fluctuations [33,34]. In the following sections, time correlation functions are considered for the HCS and use is made of stationarity and spatial homogeneity. The results must be understood as applying to system sizes for which the instability does not occur, or on time scales that are short compared to those required for growth of spatial structures.

### C. HCS averages and correlation functions

The HCS time correlation function for two phase functions  $A(\Gamma)$  and  $B(\Gamma)$  is defined as

$$C_{AB}(t, t') \equiv \langle A(t) B(t'); 0 \rangle - \langle A(t); 0 \rangle \langle B(t'); 0 \rangle, \quad (43)$$

with  $t \geq t' \geq 0$ . Here and below the brackets  $\langle ; t \rangle$  denote an average over the HCS at time  $t$ . For a system with elastic collisions in equilibrium, the above autocorrelation function can be reduced to a single time correlation function, using time translational invariance and stationarity of the Gibbs state. In the case of inelastic particles, the HCS is not stationary, but the scaling property (24) can be used to transform the correlation function to an effective time stationary average. First, use time translational invariance to write

$$\langle A(t) B(t'); 0 \rangle = \langle A(t-t') B(0); t' \rangle. \quad (44)$$

Next, transform to dimensionless variables to get

$$\begin{aligned} \langle A(t)B(t');0 \rangle &= \int d\Gamma \rho_{\text{HCS}}(\Gamma, t') A(t-t') B(0) \\ &= \int d\Gamma^* \rho_{\text{HCS}}^*(\Gamma^*) \\ &\quad \times [e^{v_c(t')(t-t')L^*} A(\{\ell \mathbf{q}_i^*, v(t') \mathbf{v}_i^*\})] \\ &\quad \times B(\{\ell \mathbf{q}_i^*, v(t') \mathbf{v}_i^*\}), \end{aligned} \quad (45)$$

where Eq. (33) has been used. Next note the identity

$$\begin{aligned} e^{-1/2\zeta^* s(t,t') \mathcal{K}^*} F(\{\mathbf{v}_i^*\}) &= F(\{e^{-1/2\zeta^* s(t,t') \mathcal{K}^*} \mathbf{v}_i^*\}) \\ &= F\left(\left\{\frac{v(t)}{v(t')} \mathbf{v}_i^*\right\}\right). \end{aligned} \quad (46)$$

The correlation function now can be written

$$\begin{aligned} \langle A(t)B(t');0 \rangle &= \int d\Gamma^* \rho_{\text{HCS}}^*(\Gamma^*) \\ &\quad \times [U(t,t') A(\{\ell \mathbf{q}_i^*, v(t) \mathbf{v}_i^*\})] \\ &\quad \times B(\ell \mathbf{q}_i^*, v(t') \mathbf{v}_i^*), \end{aligned} \quad (47)$$

where

$$U(t,t') = e^{v_c(t')(t-t')L^*} e^{1/2\zeta^* s(t,t') \mathcal{K}^*}. \quad (48)$$

This time evolution operator can be identified by differentiating with respect to  $s(t,t')$ , taking into account that in the HCS  $\zeta^*$  is time independent,

$$\begin{aligned} \frac{\partial U(t,t')}{\partial s} &= U(t,t') \left[ \frac{\partial t}{\partial s} e^{-1/2\zeta^* s(t,t') \mathcal{K}^*} v_c(t') L^* e^{1/2\zeta^* s(t,t') \mathcal{K}^*} \right. \\ &\quad \left. + \frac{1}{2} \zeta^* \mathcal{K}^* \right] \\ &= U(t,t') \left[ \frac{v_c(t')}{v_c(t)} e^{-1/2\zeta^* s(t,t') L^*} + \frac{1}{2} \zeta^* \mathcal{K}^* \right] \\ &= U(t,t') \mathcal{L}^*. \end{aligned} \quad (49)$$

Consequently,

$$U(t,t') = e^{s(t,t') \mathcal{L}^*}. \quad (50)$$

Note that this propagator is the same as that in Eq. (41) specialized to the HCS. In this case  $\zeta^*$  becomes time independent, allowing the simple exponential representation. The correlation function now can be written in the final form

$$\begin{aligned} \langle A(t)B(t');0 \rangle &= \int d\Gamma^* \rho_{\text{HCS}}^*(\Gamma^*) \\ &\quad \times [e^{s(t,t') \mathcal{L}^*} A(\{\ell \mathbf{q}_i^*, v(t) \mathbf{v}_i^*\})] \\ &\quad \times B(\ell \mathbf{q}_i^*, v(t') \mathbf{v}_i^*). \end{aligned} \quad (51)$$

This is a primary result of this section. The time correlation functions depend on the dynamics through the collision number  $s(t,t')$ . All additional time dependence occurs trivially through the thermal velocity. This is most evident when  $A$  and  $B$  are homogeneous functions of the velocity,

$$\begin{aligned} A(\{\ell \mathbf{q}_i^*, v(t) \mathbf{v}_i^*\}) &= v^a(t) A(\{\ell \mathbf{q}_i^*, \mathbf{v}_i^*\}), \\ B(\ell \mathbf{q}_i^*, v(t') \mathbf{v}_i^*) &= v^b(t') B(\{\ell \mathbf{q}_i^*, \mathbf{v}_i^*\}). \end{aligned} \quad (52)$$

Then the correlation function becomes

$$\langle A(t)B(t');0 \rangle = v^a(t) v^b(t') \langle A(s)B \rangle^*, \quad (53)$$

$$\begin{aligned} \langle A(s)B \rangle^* &= \int d\Gamma^* \rho_{\text{HCS}}^*(\Gamma^*) A(\{\ell \mathbf{q}_i^*, \mathbf{v}_i^*\}, s) \\ &\quad \times B(\{\ell \mathbf{q}_i^*, \mathbf{v}_i^*\}), \end{aligned} \quad (54)$$

and the phase function  $A(\{\ell \mathbf{q}_i^*, \mathbf{v}_i^*\}, s)$  is

$$A(\{\ell \mathbf{q}_i^*, \mathbf{v}_i^*\}, s) = e^{s(t,t') \mathcal{L}^*} A(\{\ell \mathbf{q}_i^*, \mathbf{v}_i^*\}). \quad (55)$$

This stationary-state representation for the time correlation functions simplifies considerably the analysis of response functions in the next section.

### III. IMPURITY PARTICLE DIFFUSION

In this section, the diffusion equation and associated expressions for the diffusion coefficient are derived for a granular system in the HCS. The probability density  $P(\mathbf{r}, t)$  to find the tagged or impurity particle at point  $\mathbf{r}$  at time  $t$ , given it was at the origin at  $t=0$ , is defined by

$$\begin{aligned} P(\mathbf{r}, t) &= V \langle \delta[\mathbf{q}_0(t) - \mathbf{r}] \delta(\mathbf{q}_0); 0 \rangle \\ &= \langle \delta[\mathbf{q}_0(t) - \mathbf{q}_0 - \mathbf{r}]; 0 \rangle, \end{aligned} \quad (56)$$

where the angular brackets indicate as above an average over an initial HCS and  $V$  is the volume (for  $d=3$ ) or surface (for  $d=2$ ) of the system. The second equality is a consequence of the translational invariance of the HCS. The conservation law for probability follows by differentiation of Eq. (56) with respect to  $t$ ,

$$\partial_t P(\mathbf{r}, t) + \nabla \cdot \mathbf{J}(\mathbf{r}, t) = 0, \quad (57)$$

with the probability flux  $\mathbf{J}(\mathbf{r}, t)$  identified as

$$\mathbf{J}(\mathbf{r}, t) = \langle \mathbf{v}_0(t) \delta[\mathbf{q}_0(t) - \mathbf{q}_0 - \mathbf{r}]; 0 \rangle. \quad (58)$$

The interest here is in the limiting behavior of Eq. (57) in the hydrodynamic regime, which corresponds to the long wavelength region. The long wavelength spatial dependence of  $\mathbf{J}(\mathbf{r}, t)$  can be obtained from a Fourier representation of  $P(\mathbf{r}, t)$ ,

$$\tilde{P}(\mathbf{k}, t) = \int d\mathbf{r} e^{i\mathbf{k} \cdot \mathbf{r}} P(\mathbf{r}, t). \quad (59)$$

To get a formal equation for  $\tilde{P}(\mathbf{k}, t)$ , it is useful to introduce the index of the distribution  $\tilde{C}(\mathbf{k}, t)$  by

$$\tilde{P}(\mathbf{k}, t) = e^{\tilde{C}(\mathbf{k}, t)}, \quad (60)$$

i.e.,

$$\tilde{C}(\mathbf{k}, t) = \ln \langle e^{i\mathbf{k} \cdot [\mathbf{q}_0(t) - \mathbf{q}_0(0)]}; 0 \rangle. \quad (61)$$

Differentiation with respect to time of Eq. (60) yields

$$[\partial_t - \dot{\tilde{C}}(\mathbf{k}, t)] \tilde{P}(\mathbf{k}, t) = 0, \quad (62)$$

where the dot over  $\tilde{C}$  denotes the derivative with respect to time. For long wavelengths ( $k\ell \ll 1$ ),  $\dot{\tilde{C}}(\mathbf{k}, t)$  can be expanded to order  $k^2$ ,

$$\begin{aligned} \dot{\tilde{C}}(\mathbf{k}, t) &= \frac{\langle i\mathbf{k} \cdot \mathbf{v}_0(t) e^{i\mathbf{k} \cdot [\mathbf{q}_0(t) - \mathbf{q}_0(0)]}; 0 \rangle}{\langle e^{i\mathbf{k} \cdot [\mathbf{q}_0(t) - \mathbf{q}_0(0)]}; 0 \rangle} \\ &\approx -\frac{k^2}{d} \langle \mathbf{v}_0(t) \cdot [\mathbf{q}_0(t) - \mathbf{q}_0(0)]; 0 \rangle. \end{aligned} \quad (63)$$

Substitution of this expression into Eq. (62) and inverting the transform gives Eq. (57) with the identification of the probability flux as

$$\mathbf{J}(\mathbf{r}, t) = -D(t) \nabla P(\mathbf{r}, t). \quad (64)$$

The finite time diffusion coefficient is

$$D(t) = \frac{1}{2d} \frac{\partial}{\partial t} \langle |\mathbf{q}_0(t) - \mathbf{q}_0(0)|^2; 0 \rangle. \quad (65)$$

This will be referred to as the Einstein form, relating the diffusion coefficient to the mean-square displacement. The equivalent Green-Kubo form, in terms of the velocity auto-correlation function (VACF), is derived by using the relationship

$$\mathbf{q}_0(t) - \mathbf{q}_0(0) = \int_0^t dt' \mathbf{v}_0(t'), \quad (66)$$

with the result

$$D(t) = \frac{1}{d} \int_0^t dt' \langle \mathbf{v}_0(t) \cdot \mathbf{v}_0(t'); 0 \rangle. \quad (67)$$

For normal fluids with elastic collisions, the diffusion constant follows from the long-time limit  $D = \lim_{t \rightarrow \infty} D(t)$ , or equivalently from the coefficient of the mean-square displacement when it becomes linear in  $t$ . This limit occurs for times large compared to the mean free time. Since the latter is time dependent in the HCS, the usual conditions to establish a diffusion constant, and consequently the diffusion equation, must be modified for granular media. This is done by introducing the dimensionless diffusion coefficient

$$D^*(s) = \frac{D(t)}{\ell^2 \nu_c(t)}. \quad (68)$$

Using the representation (53) for the correlation function in Eq. (67) gives the Green-Kubo form as a stationary average

$$\begin{aligned} D^*(s) &= \frac{1}{d \ell^2 \nu_c(t)} \int_0^t dt' v(t) v(t') \langle \mathbf{v}_0^*(s) \cdot \mathbf{v}_0^* \rangle^* \\ &= \frac{1}{d} \int_0^t dt' \nu_c(t') \langle \mathbf{v}_0^*(s-s') \cdot \mathbf{v}_0^* \rangle^* \\ &= \frac{1}{d} \int_0^s ds' \langle \mathbf{v}_0^*(s-s') \cdot \mathbf{v}_0^* \rangle^* \\ &= \frac{1}{d} \int_0^s ds' \langle \mathbf{v}_0^*(s') \cdot \mathbf{v}_0^* \rangle^*. \end{aligned} \quad (69)$$

In going from the first line to the second line we have written  $s(t, t') = s(t, 0) - s(t', 0) \equiv s - s'$ . Similarly, from Eqs. (65) the corresponding Einstein form is

$$D^*(s) = \frac{1}{2d} \frac{\partial}{\partial s} \langle |\mathbf{q}_0^*(s) - \mathbf{q}_0^*(0)|^2 \rangle^*. \quad (70)$$

These are the stationary average representations for the diffusion coefficient, and are the primary results of this section. In terms of the time scale  $s$ , the mean-square displacement is expected to become linear for  $s \gg 1$ , and the velocity auto-correlation function is expected to decay to zero also on this time scale. The physical interpretation of this limit is the same as for elastic collisions, since  $s$  is essentially the number of collisions per particle. However, due to the time dependence of the collision frequency, the correlation functions are expected to have the proper behavior with respect to  $s$  rather than  $t$ . This will be shown more explicitly in the next section. The dimensionless form of Eqs. (57) and (64) reads

$$\partial_s P^*(\mathbf{r}^*, s) - D^*(s) \nabla^2 P^*(\mathbf{r}^*, s) = 0, \quad (71)$$

where  $P^*(\mathbf{r}^*, s) = \ell^d P(\mathbf{r}, t)$ . Clearly, this becomes the usual diffusion equation for sufficiently large  $s$ , if  $D^*(s)$  tends to a constant.

It is useful to introduce a dimensionless VACF for the impurity particle that is normalized to unity at  $s=0$ . This requires calculation of  $\langle v_0^{*2} \rangle^*$ . It might appear from Eq. (20) that this can be obtained simply in terms of the mass of the impurity and the temperature of the fluid. However, it has been shown elsewhere that mechanically different particles in a common HCS do not have the same temperatures [26], as already mentioned below Eq. (25). Thus  $\langle v_0^{*2} \rangle^*$  is given by Eq. (20) with both  $m$  and  $T(t)$  replaced by  $m_0$  and  $T_0(t)$ ,

$$\langle v_0^{*2} \rangle^* = \frac{dT_0(t)}{m_0 v^2(t)} = \frac{d}{2} \phi_{\text{HCS}}, \quad (72)$$

where  $\phi_{\text{HCS}}$  is the ratio of the square of the thermal velocity for the impurity particle relative to that for the fluid particles,

$$\phi_{\text{HCS}} = \frac{mT_0(t)}{m_0T(t)}. \quad (73)$$

Since the cooling rates of the fluid,  $\zeta(t)$ , and the impurity particle,  $\zeta_0(t)$ , are the same and they are proportional to the square root of the temperature, the above ratio is time independent. The condition for equal cooling rates also determines  $\phi_{\text{HCS}}$ . These rates are calculated to good approximation using a local equilibrium ensemble in Appendix A, and are given by

$$\zeta^* = \frac{2^{1/2}\pi^{(d-1)/2}}{\Gamma(d/2)d} \chi(1-\alpha^2), \quad (74)$$

$$\zeta_0^* = \nu^* \left( 1 - h \frac{1 + \phi_{\text{HCS}}}{\phi_{\text{HCS}}} \right) (1 + \phi_{\text{HCS}})^{1/2}, \quad (75)$$

where  $h = (1 + \alpha_0)m/2(m + m_0)$ , and  $\nu^*$  is a dimensionless impurity particle collision rate,

$$\nu^* = \frac{8h}{d} \left( \frac{\bar{\sigma}}{\sigma} \right)^{d-1} \frac{\pi^{(d-1)/2}}{\Gamma(d/2)} \chi_0. \quad (76)$$

The factors  $\chi$  and  $\chi_0$  are the pair-correlation function for the fluid-fluid and the fluid-impurity particles at contact, respectively. A more accurate calculation for the case of hard spheres ( $d=3$ ) is carried out in Ref. [27]. Equating Eqs. (74) and (75) provides the equation for  $\phi_{\text{HCS}}$ ,

$$(1 + \phi_{\text{HCS}})^{1/2} \left( 1 - h \frac{1 + \phi_{\text{HCS}}}{\phi_{\text{HCS}}} \right) = \frac{\zeta^*}{\nu^*}. \quad (77)$$

This gives a cubic equation which has a unique real, positive solution for all allowed values of  $h$  and  $\zeta^*/\nu^*$ . For elastic collisions,  $\phi_{\text{HCS}} \rightarrow m/m_0$  as required by the equipartition theorem. Qualitative changes in this solution, similar to a phase transition occur in the limit  $h \rightarrow 0$  [28], but will not be discussed here. The normalized VACF is now given by

$$C_{vv}^*(s) \equiv \frac{\langle \mathbf{v}_0^*(s) \cdot \mathbf{v}_0^* \rangle^*}{\langle v_0^{*2} \rangle^*} = \frac{2}{d\phi_{\text{HCS}}} \langle \mathbf{v}_0^*(s) \cdot \mathbf{v}_0^* \rangle^* \quad (78)$$

and the diffusion coefficient in Eq. (69) becomes

$$D^*(s) = \frac{\phi_{\text{HCS}}}{2} \int_0^s ds' C_{vv}^*(s'). \quad (79)$$

#### IV. APPROXIMATIONS

In the following, two approximations, originally developed for fluids with elastic collisions [35], are applied to calculate the VACF in the HCS. The first method uses a leading-order truncation of a cumulant expansion, while the second uses an approximate kinetic equation. The results confirm the expected time scale for transition to hydrodynamics and provide the detailed dependence on density and restitution coefficients.

#### A. Cumulant expansion

The cumulant expansion of the VACF is

$$C_{vv}^*(s) = \exp \left[ \sum_{p=1}^{\infty} \frac{1}{p!} \omega_p^* (-s)^p \right]. \quad (80)$$

The coefficients  $\omega_p^*$  are determined from the initial time derivatives of the correlation function. Clearly, truncation of this expansion at any order is asymptotically exact at short times and also for small  $h$  (heavy impurity) since each time derivative contributes a factor of  $h$ . The simplest such approximation retains only the leading term

$$C_{vv}^*(s) \approx e^{-\omega_1^* s}, \quad (81)$$

with

$$\begin{aligned} \omega_1^* &= - \left[ \frac{\partial}{\partial s} \ln C_{vv}^*(s) \right]_{s=0} \\ &= - \frac{2}{d\phi_{\text{HCS}}} \langle (\mathcal{L}^* \mathbf{v}_0^* \cdot \mathbf{v}_0^*)^* \rangle. \end{aligned} \quad (82)$$

Use has been made of the definition of the  $s$  dependence in Eq. (78). The corresponding approximation for the mean-square displacement is obtained by integrating Eq. (81) twice,

$$\langle |\mathbf{q}_0^*(s) - \mathbf{q}_0^*(0)|^2 \rangle^* = \frac{d\phi_{\text{HCS}}}{\omega_1^*} [s - \omega_1^*^{-1} (1 - e^{-\omega_1^* s})]. \quad (83)$$

For elastic collisions, this approximation is known to be accurate for short as well as long times, and for a wide range of densities and mass ratios. The resulting diffusion coefficient and diffusion constant are then found by substituting Eq. (81) into Eq. (79),

$$D^*(s) = D^* (1 - e^{-\omega_1^* s}), \quad (84)$$

$$D^* = \lim_{s \rightarrow \infty} D^*(s) = \frac{\phi_{\text{HCS}}}{2\omega_1^*}. \quad (85)$$

Clearly,  $\omega_1^*$  is a characteristic dimensionless collision frequency for the impurity particle. The first cumulant approximation confirms the expectation that the mean-square displacement becomes linear in  $s$  and the velocity autocorrelation function decays for  $s \gg \omega_1^*$ . Consequently, the macroscopic diffusion equation applies on this time scale as well. The collision frequency  $\omega_1^*$  is evaluated in Appendix A by using a local equilibrium ensemble, with the result

$$\omega_1^* = -\frac{1}{2} \zeta^* + \frac{1}{2} \nu^* (1 + \phi_{\text{HCS}})^{1/2}, \quad (86)$$

where  $\zeta^*$  and  $\nu^*$  are defined in Eqs. (74) and (76), respectively. Substitution of this into Eq. (85) gives



$$D^* = \frac{\phi_{\text{HCS}}}{(1 + \phi_{\text{HCS}})^{1/2} \nu^* - \zeta^*}. \quad (87)$$

It is possible to show that  $D^*$  is positive and finite for all values of the density and restitution coefficients.

The failure of energy equipartition means that  $\phi_{\text{HCS}}$  is not simply the mass ratio  $m/m_0$  but changes also with the restitution coefficients and the size ratio. To illustrate this, use Eq. (77) to write Eq. (87) in the equivalent form

$$D^* \nu^* = \frac{\phi_{\text{HCS}}^2}{h(1 + \phi_{\text{HCS}})^{3/2}}. \quad (88)$$

Let the mass ratio be fixed. The right side of Eq. (88) has a minimum when the left side of Eq. (77) vanishes, i.e.,  $\phi_{\text{HCS}} \rightarrow h/(1-h)$ . This is a possible solution for mechanically different particles only if  $\zeta^*/\nu^* \rightarrow 0$ , i.e., elastic collisions among the fluid particles. Thus,  $D^* \nu^*$  is smallest when the fluid cooling rate is small compared to the impurity collision rate. Conversely, the maximum value of  $D^* \nu^*$  occurs at the maximum value for  $\phi_{\text{HCS}} \rightarrow [\zeta^*/\nu^*(1-h)]^2$ . This occurs when the cooling rate is large compared to the impurity collision rate, which is possible when the mechanical differences (e.g., mass ratio, size ratio) are large.

### B. Kinetic theory

Perhaps the most accurate and detailed evaluation of time correlation functions is via kinetic theory methods. These can be applied as well to the case of inelastic collisions [30,36,17]. To show this, first use the adjoint property of the Liouville operators to write the velocity autocorrelation function in the form

$$\begin{aligned} C_{vv}^*(s) &= \frac{2}{d\phi_{\text{HCS}}} \int d\Gamma^* \rho_{\text{HCS}}^*(\Gamma^*) \mathbf{v}_0^* \cdot e^{s\bar{L}^*} \mathbf{v}_0^* \\ &= \frac{2}{d\phi_{\text{HCS}}} \int d\Gamma^* \mathbf{v}_0^* \cdot e^{-s\bar{L}^*} [\rho_{\text{HCS}}^*(\Gamma^*) \mathbf{v}_0^*]. \end{aligned} \quad (89)$$

Next, define the dimensionless reduced correlation functions in a manner similar to Eq. (19)

$$\begin{aligned} \psi^{*(l+1)}(x_0^*, \dots, x_l^*, s) &\equiv N^l \int dx_{l+1}^* \dots dx_N^* e^{-s\bar{L}^*} \\ &\times [\rho_{\text{HCS}}^*(\Gamma^*) \mathbf{v}_0^*], \end{aligned} \quad (90)$$

so that the VACF can be written as

$$C_{vv}^*(s) = \frac{2}{d\phi_{\text{HCS}}} \int dx_0^* \mathbf{v}_0^* \cdot \psi^{*(1)}(x_0^*, s). \quad (91)$$

It is easily verified by direct differentiation that the  $\psi^{*(l+1)}$  functions obey a hierarchy of equations similar to Eq. (18), the first of which is

$$\begin{aligned} (\partial_s + \mathbf{v}_0^* \cdot \nabla_0^*) \psi^{*(1)}(x_0^*, s) + \frac{1}{2} \zeta^* \frac{\partial}{\partial \mathbf{v}_0^*} \cdot [\mathbf{v}_0^* \psi^{*(1)}(x_0^*, s)] \\ = \int dx_1^* \bar{T}^*(0,1) \psi^{*(2)}(x_0^*, x_1^*, s). \end{aligned} \quad (92)$$

A kinetic equation results from a closure approximation in the above equation that expresses  $\psi^{*(2)}$  as a functional of  $\psi^{*(1)}$ . Formally, this is possible since both  $\psi^{*(1)}(x_0^*, s)$  and  $\psi^{*(2)}(x_0^*, x_1^*, s)$  are linear functionals of  $\psi^{*(1)}(x_0^*, 0)$ . In principle, this functional relationship can be inverted to give  $\psi^{*(2)}(x_0^*, x_1^*, s) = \Psi^{*(2)}[x_0^*, x_1^*, s | \psi^{*(1)}]$ . Use of this in Eq. (92) provides the closed kinetic equation for  $\psi^{*(1)}$ . In practice, it is a difficult many-body problem to discover this functional. However, its form is easily calculated at  $s=0$ . Equation (90) gives

$$\begin{aligned} \Psi^{*(2)}[x_0^*, x_1^*, s=0 | \psi^{*(1)}(0)] \\ = f^{*(2)}(x_0^*, x_1^*) \mathbf{v}_0^* \\ = f^{*(2)}(x_0^*, x_1^*) [f^{*(1)}(x_0^*)]^{-1} \psi^{*(1)}(x_0^*, 0), \end{aligned} \quad (93)$$

where  $f^{*(1)}(x_0^*)$  and  $f^{*(2)}(x_0^*, x_1^*)$  are the reduced distribution functions associated with  $\rho_{\text{HCS}}^*(\Gamma^*)$ . For fluids with elastic collisions, the approximation

$$\Psi^{*(2)}[x_0^*, x_1^*, s | \psi^{*(1)}] \rightarrow \Psi^{*(2)}[x_0^*, x_1^*, 0 | \psi^{*(1)}(s)] \quad (94)$$

is accurate over a wide range of low-to-moderate densities. The same approximation in the current case leads to the kinetic equation

$$\begin{aligned} (\partial_s + \mathbf{v}_0^* \cdot \nabla_0^*) \psi^{*(1)}(x_0^*, s) + \frac{\zeta^*}{2} \frac{\partial}{\partial \mathbf{v}_0^*} \cdot [\mathbf{v}_0^* \psi^{*(1)}(x_0^*, s)] \\ = \int dx_1^* \bar{T}^*(0,1) f^{*(2)}(x_0^*, x_1^*) \\ \times [f^{*(1)}(x_0^*)]^{-1} \psi^{*(1)}(x_0^*, s). \end{aligned} \quad (95)$$

This equation is exact at short times by construction and its use for longer times can be interpreted as a Markovian approximation [37]. For the VACF only the spatial integral of  $\psi^{*(1)}(x_0^*, s)$  is required, so the final representation of Eq. (91) becomes

$$C_{vv}^*(s) = \frac{2}{d\phi_{\text{HCS}}} \int d\mathbf{v}_0^* \mathbf{v}_0^* \cdot \psi^*(\mathbf{v}_0^*, s), \quad (96)$$

where

$$\psi^*(\mathbf{v}_0^*, s) = \int d\mathbf{q}_0^* \psi^{*(1)}(x_0^*, s) \quad (97)$$

obeys the kinetic equation

$$\partial_s \psi^*(\mathbf{v}_0^*, s) + \frac{\zeta^*}{2} \frac{\partial}{\partial \mathbf{v}_0^*} \cdot [\mathbf{v}_0^* \psi^*(\mathbf{v}_0^*, s)] = \mathcal{I} \psi^*(\mathbf{v}_0^*, s). \quad (98)$$

The collision operator  $\mathcal{I}$  is defined as

$$\begin{aligned} \mathcal{I} \psi^*(\mathbf{v}_0^*, s) \equiv & \int dx_1^* \bar{T}^*(0,1) f^{*(2)}(x_0^*, x_1^*) \\ & \times [f^{*(1)}(x_0^*)]^{-1} \psi(\mathbf{v}_0^*, s). \end{aligned} \quad (99)$$

The kinetic Eq. (98) has to be solved with the initial condition

$$\psi^*(\mathbf{v}_0^*, 0) = f^{*(1)}(\mathbf{v}_0^*) \mathbf{v}_0^*, \quad (100)$$

as follows directly from Eq. (90). To obtain Eq. (98) from Eq. (95), use has been made of the fact that the HCS distributions are spatially homogeneous. For  $\alpha=1$  the linear collision operator  $\mathcal{I}$  is non-negative, and the correlation function is found to decay on a time scale of the order of  $s \sim \omega_1^{*-1}$ , the initial rate of decay. Although not proven, it is reasonable to assume that the spectrum of  $\mathcal{I}$  is qualitatively similar for  $\alpha < 1$ . Since the kinetic equation is exact at  $s \rightarrow 0$ , the leading term in a cumulant expansion of Eq. (90) also is exact and agrees with Eq. (81). More generally, this approximate kinetic equation gives contributions to all higher terms in the cumulant expansion. However, for  $\alpha=1$  these corrections are of the order of a few percent except when the size or mass ratio of fluid and impurity particles differs greatly from one.

The diffusion constant is obtained by integrating Eq. (91) to get

$$D^* = - \frac{2}{d \phi_{\text{HCS}}} \int d\mathbf{v}_0^* \mathbf{v}_0^* \cdot \mathbf{X}(\mathbf{v}_0^*), \quad (101)$$

where  $\mathbf{X}(\mathbf{v}_0^*)$  is the solution to the integral equation

$$\mathcal{I} X_i(\mathbf{v}_0^*) - \frac{\zeta^*}{2} \frac{\partial}{\partial \mathbf{v}_0^*} \cdot [\mathbf{v}_0^* X_i(\mathbf{v}_0^*)] = f^{*(1)}(\mathbf{v}_0^*) v_{0i}^*. \quad (102)$$

The only (left) eigenfunction with vanishing eigenvalue of the operator defined by the left-hand side of the above equation is one. The right side is orthogonal to one so the Fredholm alternative (solubility condition) is satisfied and a solution to this equation exists. It is shown in Appendix A that the diffusion coefficient given by Eq. (101) is the same as that obtained from the Chapman-Enskog solution to the Enskog kinetic equation, if velocity correlations are neglected for  $f^{*(2)}(x_0^*, x_1^*)$  in the definition of  $\mathcal{I}$ , Eq. (99). Recent computer simulations confirm that such neglect is a good approximation [38]. Finally, if Eq. (102) is solved as an expansion in Sonine polynomials (the usual method for elastic collisions), the first approximation yields again the leading cumulant approximation. Thus the cumulant approximation, linear kinetic theory, and Chapman-Enskog solution to the

Enskog kinetic equation are all the same modulo small differences due to velocity correlations and higher order Sonine polynomials.

## V. DISCUSSION

In this paper, it has been shown that standard linear-response theory can be extended in a natural way to describe diffusion in a system of inelastic hard spheres in the homogeneous cooling state. The response functions of the system are given in terms of stationary-state averages corresponding to an effective dimensionless dynamics. In particular, the dimensionless time scale is related to the average number of collisions per particle taking place in the system, and it is the physically relevant one to analyze the aging to a hydrodynamic stage. Similar Einstein and Green-Kubo expressions have been obtained for the shear viscosity [39,40], which is essentially a diffusion process like that considered here. Response to an external force has been studied to determine expressions for the mobility [27]. Transport coefficients associated with longitudinal hydrodynamic modes (e.g., thermal conductivity) pose special problems and will be discussed elsewhere. Beyond their formal interest and utility for approximate analysis, the results derived here enable a systematic nonperturbative study of transport processes in granular fluids by means of molecular-dynamics simulation of the response functions, just as for fluids with elastic collisions.

It is interesting to consider the particular case of self diffusion, i.e., when the impurity particle is mechanically equivalent to the fluid particles, for which previous analysis have been carried out. The expression for the self-diffusion coefficient in the first cumulant approximation is obtained by considering the limit  $\phi_{\text{HCS}}=1$ ,  $h=(1+\alpha)/4$ , and  $\alpha_0=\alpha$  in Eq. (87). This yields

$$D^* = \frac{d\Gamma(d/2)}{(1+\alpha)^2 \chi^2 2^{1/2} \pi^{(d-1)/2}}. \quad (103)$$

As expected, this result agrees with the expression derived in Ref. [20] from a Chapman-Enskog solution to the Enskog-Lorentz equation in a first-order polynomial expansion. A previous approach to self diffusion in the HCS, also in the context of linear response theory, has been developed by Brilliantov and Pöschel [41]. Their result differs from Eq. (103) in a factor of  $(1+\alpha)/2$ . Quite peculiarly, the same result had been derived before by Hsiao and Hunt [42] and Savage and Dai [43], independently, from approximate solutions of the Enskog-Lorentz equation. The discrepancy between the result in Ref. [41] and the one reported here is not due to a different degree of approximation, but has a fundamental physical origin. Brilliantov and Pöschel assume that the velocity autocorrelation function of the tagged particle has the form

$$[C_{vv}(t, t')]_{BP} = v(t)^2 e^{-(t-t')/\tau_B(t')}, \quad (104)$$

with the relaxation time  $\tau_B$  being inversely proportional to the initial slope, in the actual time scale  $t$ , of the VACF. This

is to be contrasted with our analysis, based in the relevance of the time scale defined by the number of collisions,

$$C_{vv}(t, t') = v(t)v(t') \exp \left[ -\omega_1^* \int_{t'}^t d\tau \frac{v(\tau)}{\ell} \right], \quad (105)$$

with  $\omega_1^*$  determined from the initial slope of the VACF in the dimensionless scale defined by Eq. (27). Making clear the crucial role played by this latter time scale for the study of response functions is one of the main goals in this paper.

The quality of the simple approximations given here is studied in the following paper [29] by comparison with molecular-dynamics simulations for both the Einstein and Green-Kubo forms. Only the case of self-diffusion is considered. It is found that the agreement is very good for low densities and all degrees of dissipation, but there are large discrepancies at high density and large dissipation. The possible reasons for the discrepancies are discussed there.

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### APPENDIX A: COOLING RATES IN HCS

In this appendix, the cooling rates for the fluid and the impurity particle, as well as first cumulant for the VACF, are calculated from the Liouville dynamics of the system. For simplicity, a local equilibrium ensemble approximation, known to lead to accurate results [7], will be considered. The dimensionless cooling rates for the fluid and impurity particle are defined as

$$\zeta^* \equiv -\frac{\ell}{v(t)} \frac{\partial \ln T}{\partial t} = -\frac{2}{d} \langle L^* \mathbf{v}_1^{*2} \rangle^*, \quad (A1)$$

$$\zeta_0^* \equiv -\frac{\ell}{v(t)} \frac{\partial \ln T_0}{\partial t} = -\frac{2}{d\phi_{\text{HCS}}} \langle L^* \mathbf{v}_0^{*2} \rangle^*, \quad (A2)$$

respectively. For the derivation of the last equalities, use has been made of the property that in the HCS, for any dynamical variable  $A(\Gamma)$  having the scaling property (52) it is

$$\langle LA; t \rangle = \frac{v^{a+1}(t)}{\ell} \langle L^* A(\{\ell \mathbf{q}_i^*, \mathbf{v}_i^*\}) \rangle^*. \quad (A3)$$

The first cumulant for the VACF is given by Eq. (82) or, equivalently,

$$\omega_1^* = -\frac{\zeta^*}{2} - \frac{2}{d\phi_{\text{HCS}}} \langle (L^* \mathbf{v}_0^*) \cdot \mathbf{v}_0^* \rangle^*. \quad (A4)$$

In terms of the dimensionless distribution functions

$$f^{*(2)}(x_0^*, x_1^*) = N \int dx_2^* dx_3^* \cdots dx_N^* \rho_{\text{HCS}}^*(\Gamma^*), \quad (A5)$$

and

$$f^{*(2)}(x_1^*, x_2^*) = N^2 \int dx_0^* dx_3^* \cdots dx_N^* \rho_{\text{HCS}}^*(\Gamma^*), \quad (A6)$$

the above quantities can be written as

$$\zeta^* = -\frac{2}{dN} \int dx_1^* dx_2^* f^{*(2)}(x_1^*, x_2^*) T^*(1, 2) \mathbf{v}_1^{*2}, \quad (A7)$$

$$\zeta_0^* = -\frac{2}{d\phi_{\text{HCS}}} \int dx_0^* dx_1^* f^{*(2)}(x_0^*, x_1^*) T^*(0, 1) \mathbf{v}_0^{*2}, \quad (A8)$$

$$\omega_1^* = -\frac{\zeta^*}{2} - \frac{2}{d\phi_{\text{HCS}}} \int dx_0^* dx_1^* f^{*(2)}(x_0^*, x_1^*) \times \mathbf{v}_0^* \cdot [T^*(0, 1) \mathbf{v}_0^*]. \quad (A9)$$

The action of the binary collision operators can be performed for further simplification, leading to

$$\zeta^* = \frac{1}{2dn^*} (1 - \alpha^2) \sigma^{*d-1} \int d\mathbf{v}_1^* \int d\mathbf{v}_2^* \int d\Omega \times f^{*(2)}(\mathbf{v}_1^*, \mathbf{v}_2^*, \mathbf{q}_{12}^* = \boldsymbol{\sigma}^*) \Theta(-\mathbf{g}_{12}^* \cdot \hat{\boldsymbol{\sigma}}) |\mathbf{g}_{12}^* \cdot \hat{\boldsymbol{\sigma}}|^3, \quad (A10)$$

$$\zeta_0^* = -\frac{4h}{d\phi_{\text{HCS}}} \bar{\sigma}^{*d-1} V^* \int d\mathbf{v}_0^* \int d\mathbf{v}_1^* \int d\Omega \times f_0^{*(2)}(\mathbf{v}_0^*, \mathbf{v}_1^*, \mathbf{q}_{01}^* = \bar{\boldsymbol{\sigma}}^*) \Theta(-\mathbf{g}_{01}^* \cdot \hat{\boldsymbol{\sigma}}) |\mathbf{g}_{01}^* \cdot \hat{\boldsymbol{\sigma}}|^2 \times \left[ 2\mathbf{G}_{01}^* \cdot \hat{\boldsymbol{\sigma}} + \frac{m}{m+m_0} (1 - \alpha_0) \hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{01}^* \right], \quad (A11)$$

$$\omega_1^* = -\frac{\zeta^*}{2} - \frac{4h}{d\phi_{\text{HCS}}} \bar{\sigma}^{*d-1} V^* \int d\mathbf{v}_0^* \int d\mathbf{v}_1^* \int d\Omega \times f^{*(2)}(\mathbf{v}_0^*, \mathbf{v}_1^*, \mathbf{q}_{01}^* = \bar{\boldsymbol{\sigma}}) \Theta(-\mathbf{g}_{01}^* \cdot \hat{\boldsymbol{\sigma}}) |\mathbf{g}_{01}^* \cdot \hat{\boldsymbol{\sigma}}|^2 \times \left( \mathbf{G}_{01}^* \cdot \hat{\boldsymbol{\sigma}} + \frac{m}{m_0+m} \mathbf{g}_{01} \cdot \hat{\boldsymbol{\sigma}} \right), \quad (A12)$$

where  $h$  is defined below Eq. (75),  $V^*$  is the reduced volume or surface of the system, and  $\mathbf{G}_{01}^*$  is the reduced velocity of the center of mass for particles 0 and 1, i.e.,

$$\mathbf{G}_{01}^* = \frac{m_0 \mathbf{v}_0^* + m \mathbf{v}_1^*}{m_0 + m}. \quad (A13)$$

The above results are still exact. To compute the integrals two approximations are introduced. First, the velocity correlations in  $f^{*(2)*}(\mathbf{v}_1^*, \mathbf{v}_2^*, \mathbf{q}_{12}^* = \boldsymbol{\sigma}^*)$  and  $f_0^{*(2)}(\mathbf{v}_0^*, \mathbf{v}_1^*, \mathbf{q}_{01}^* = \bar{\boldsymbol{\sigma}}^*)$  are neglected. Note that this approximation is required only for particles at contact and for the precollision hemisphere. Significant velocity correlations exist on the op-

posite hemisphere and at larger separation of the particles, and no restriction is placed on these configurations. The second approximation is to represent the one-particle distributions by Maxwellians. This is known to be a good approximation except for conditions of large mechanical differences between the fluid and impurity particles. These two approximations are equivalent to write

$$f^{*(2)}(\mathbf{v}_0^*, \mathbf{v}_1^*, \mathbf{q}_{01}^* = \bar{\boldsymbol{\sigma}}) = n^* \chi_0(\bar{\boldsymbol{\sigma}}) \varphi_0^*(\mathbf{v}_0^*) \varphi^*(\mathbf{v}_1^*), \quad (\text{A14})$$

$$f^{*(2)}(\mathbf{v}_1^*, \mathbf{v}_2^*, \mathbf{q}_{12}^* = \boldsymbol{\sigma}) = n^{*2} \chi(\boldsymbol{\sigma}) \varphi^*(\mathbf{v}_1^*) \varphi^*(\mathbf{v}_2^*), \quad (\text{A15})$$

where  $\chi_0(\bar{\boldsymbol{\sigma}})$  and  $\chi(\boldsymbol{\sigma})$  are the fluid-impurity and fluid-fluid pair-correlation functions for particles at contact, and

$$\varphi_0^*(\mathbf{v}_0^*) = \frac{1}{(\phi_{\text{HCS}} \pi)^{d/2}} e^{-v_0^{*2}/\phi_{\text{HCS}}}, \quad (\text{A16})$$

$$\varphi^*(\mathbf{v}_i^*) = \frac{1}{\pi^{d/2}} e^{-v_i^{*2}}. \quad (\text{A17})$$

Now the integrations in Eqs. (A10)–(A12) can be performed. The calculations are straightforward but lengthy, and will be not reproduce here. The results are given by Eqs. (74)–(76), and (86).

## APPENDIX B: ENSKOG KINETIC THEORY

In the main text sections, linear response has been applied to describe diffusion directly from the Liouville dynamics. It is useful to see that equivalent results, in appropriate approximations, follow from kinetic theory as well. This is illustrated in this appendix using the Enskog kinetic theory, expected to be valid over a wide range of densities and restitution coefficients. To review its origin and applicability to granular fluids, the first hierarchy equations of Eq. (18) for the impurity particle reduced distribution function  $f_0^{(1)}(\mathbf{q}_0, \mathbf{v}_0, t)$  and one-particle fluid reduced distribution function  $f^{(1)}(\mathbf{q}_1, \mathbf{v}_1, t)$ , are written explicitly,

$$\begin{aligned} (\partial_t + \mathbf{v}_0 \cdot \nabla_0) f_0^{(1)}(\mathbf{q}_0, \mathbf{v}_0, t) &= \bar{\sigma}^{d-1} \int d\mathbf{q}_1 \int d\mathbf{v}_1 \int d\Omega \Theta(-\mathbf{g}_{01} \cdot \hat{\boldsymbol{\sigma}}) |\mathbf{g}_{01} \cdot \hat{\boldsymbol{\sigma}}| \\ &\times [\alpha_0^{-2} \delta(\mathbf{q}_{01} - \bar{\boldsymbol{\sigma}}) b_{01}^{-1} - \delta(\mathbf{q}_{01} + \bar{\boldsymbol{\sigma}})] \\ &\times f^{(2)}(\mathbf{q}_0, \mathbf{v}_0, \mathbf{q}_1, \mathbf{v}_1, t), \end{aligned} \quad (\text{B1})$$

$$\begin{aligned} (\partial_t + \mathbf{v}_1 \cdot \nabla_1) f^{(1)}(\mathbf{q}_1, \mathbf{v}_1, t) &= \sigma^{d-1} \int d\mathbf{q}_2 \int d\mathbf{v}_2 \int d\Omega \Theta(-\mathbf{g}_{12} \cdot \hat{\boldsymbol{\sigma}}) |\mathbf{g}_{12} \cdot \hat{\boldsymbol{\sigma}}| \\ &\times [\alpha^{-2} \delta(\mathbf{q}_{12} - \boldsymbol{\sigma}) b_{12}^{-1} - \delta(\mathbf{q}_{12} + \boldsymbol{\sigma})] \\ &\times f^{(2)}(\mathbf{q}_1, \mathbf{v}_1, \mathbf{q}_2, \mathbf{v}_2, t), \end{aligned} \quad (\text{B2})$$

where  $f^{(2)}(\mathbf{q}_0, \mathbf{v}_0, \mathbf{q}_1, \mathbf{v}_1, t)$  and  $f^{(2)}(\mathbf{q}_1, \mathbf{v}_1, \mathbf{q}_2, \mathbf{v}_2, t)$  are the two-particle reduced distribution functions for the impurity and one-fluid particle, and for two-fluid particles, respectively. Their definitions are given by Eqs. (19) and (23). A closure of the hierarchy is obtained by replacing in the above,

$$f^{(2)}(\mathbf{q}_0, \mathbf{v}_0, \mathbf{q}_1, \mathbf{v}_1, t) \rightarrow \chi_0(\mathbf{q}_0, \mathbf{q}_1; t) \times f_0^{(1)}(\mathbf{q}_0, \mathbf{v}_0, t) f^{(1)}(\mathbf{q}_1, \mathbf{v}_1, t), \quad (\text{B3})$$

$$f^{(2)}(\mathbf{q}_1, \mathbf{v}_1, \mathbf{q}_2, \mathbf{v}_2, t) \rightarrow \chi(\mathbf{q}_1, \mathbf{q}_2; t) \times f^{(1)}(\mathbf{q}_1, \mathbf{v}_1, t) f^{(1)}(\mathbf{q}_2, \mathbf{v}_2, t). \quad (\text{B4})$$

The approximation is a generalization of that in Eq. (94), and also of that in Eqs. (A14) and (A15). It can be understood in two different ways. It is the exact Markovian limit if the initial distribution functions have the forms (B3) and (B4) on the precollisional hemisphere (exact at short times). It also follows if velocity correlations on this hemisphere are destroyed between collisions (Boltzmann's argument). In any case, it is known to provide a good description of the hard-sphere fluid over a wide range of densities and times for elastic collisions, and this validity appears to hold as well for inelastic collisions. As in the elastic collisions case, the functions  $\chi_0(\mathbf{q}_0, \mathbf{q}_1; t)$  and  $\chi(\mathbf{q}_1, \mathbf{q}_2; t)$  are taken to be the equilibrium configurational pair-correlation functions as functionals of the nonequilibrium density.

The Enskog approximation converts Eqs. (B3) and (B4) into a pair of kinetic equations. The kinetic equation for the fluid is autonomous while that for the impurity distribution is a functional of  $f^{(1)}(\mathbf{q}_1, \mathbf{v}_1, t)$ ,

$$(\partial_t + \mathbf{v}_1 \cdot \nabla_1) f(\mathbf{q}_1, \mathbf{v}_1, t) = J_E[\mathbf{q}_1, \mathbf{v}_1 | f(t)], \quad (\text{B5})$$

$$(\partial_t + \mathbf{v}_0 \cdot \nabla_0) f_0(\mathbf{q}_0, \mathbf{v}_0, t) = J_{\text{EL}}[\mathbf{q}_0, \mathbf{v}_0 | f_0(t), f(t)]. \quad (\text{B6})$$

Here and below the superscript on the single-particle distributions will be suppressed for simplicity of notation. The Enskog and Enskog-Lorentz collision operators are given by

$$\begin{aligned} J_E[\mathbf{q}_1, \mathbf{v}_1 | f(t)] &\equiv \sigma^{d-1} \int d\mathbf{v}_2 \int d\Omega \Theta(-\mathbf{g}_{12} \cdot \hat{\boldsymbol{\sigma}}) |\mathbf{g}_{12} \cdot \hat{\boldsymbol{\sigma}}| \\ &\times \{ \alpha^{-2} \chi[\mathbf{q}_1, \mathbf{q}_1 - \boldsymbol{\sigma} | n(t)] b_{12}^{-1} f(\mathbf{q}_1, \mathbf{v}_1, t) \\ &\times f(\mathbf{q}_1 - \boldsymbol{\sigma}, \mathbf{v}_2, t) - \chi[\mathbf{q}_1, \mathbf{q}_1 + \boldsymbol{\sigma} | n(t)] f(\mathbf{q}_1, \mathbf{v}_1, t) \\ &\times f(\mathbf{q}_1 + \boldsymbol{\sigma}, \mathbf{v}_2, t) \}, \end{aligned} \quad (\text{B7})$$

$$\begin{aligned} J_{\text{EL}}[\mathbf{q}_0, \mathbf{v}_0 | f_0(t), f(t)] &\equiv \bar{\sigma}^{d-1} \int d\mathbf{v}_1 \int d\Omega \Theta(-\mathbf{g}_{01} \cdot \hat{\boldsymbol{\sigma}}) |\mathbf{g}_{01} \cdot \hat{\boldsymbol{\sigma}}| \\ &\times \{ \alpha_0^{-2} \chi_0[\mathbf{q}_0, \mathbf{q}_0 - \bar{\boldsymbol{\sigma}} | n(t)] b_{01}^{-1} f_0(\mathbf{q}_0, \mathbf{v}_0, t) \\ &\times f(\mathbf{q}_0 - \bar{\boldsymbol{\sigma}}, \mathbf{v}_1, t) - \chi_0[\mathbf{q}_0, \mathbf{q}_0 + \bar{\boldsymbol{\sigma}} | n(t)] f_0(\mathbf{q}_0, \mathbf{v}_0, t) \\ &\times f(\mathbf{q}_0 + \bar{\boldsymbol{\sigma}}, \mathbf{v}_1, t) \}, \end{aligned} \quad (\text{B8})$$

respectively. The HCS is a special case for which Eqs. (B5) and (B6) reduce to

$$\partial_t f_{\text{HCS}} = J_E[\mathbf{v}_1 | f_{\text{HCS}}(t)], \quad (\text{B9})$$

$$\partial_t f_{0,\text{HCS}} = J_{\text{EL}}[\mathbf{v}_0 | f_{0,\text{HCS}}(t), f_{\text{HCS}}(t)], \quad (\text{B10})$$

with the collision terms

$$\begin{aligned} & J_E[\mathbf{v}_1 | f_{\text{HCS}}(t)] \\ & \equiv \sigma^{d-1} \chi \int d\mathbf{v}_2 \int d\Omega \Theta(-\mathbf{g}_{12} \cdot \hat{\boldsymbol{\sigma}}) |\mathbf{g}_{12} \cdot \hat{\boldsymbol{\sigma}}| \\ & \quad \times [(\alpha^{-2} b_{12}^{-1} - 1) f_{\text{HCS}}(\mathbf{v}_1, t) f_{\text{HCS}}(\mathbf{v}_2, t)], \end{aligned} \quad (\text{B11})$$

$$\begin{aligned} & J_{\text{EL}}[\mathbf{v}_0 | f_{0,\text{HCS}}(t), f_{\text{HCS}}(t)] \\ & \equiv \bar{\sigma}^{d-1} \chi_0 \int d\mathbf{v}_1 \int d\Omega \Theta(-\mathbf{g}_{01} \cdot \hat{\boldsymbol{\sigma}}) \\ & \quad \times |\mathbf{g}_{01} \cdot \hat{\boldsymbol{\sigma}}| (\alpha_0^{-2} b_{01}^{-1} - 1) f_{0,\text{HCS}}(\mathbf{v}_0, t) f_{\text{HCS}}(\mathbf{v}_1, t). \end{aligned} \quad (\text{B12})$$

Use has been made of the fact that  $\chi[\mathbf{q}_1, \mathbf{q}_1 - \boldsymbol{\sigma} | n] \equiv \chi$  and  $\chi_0[\mathbf{q}_0, \mathbf{q}_0 - \boldsymbol{\sigma} | n] \equiv \chi_0$  are independent of space coordinates due to translational invariance. Furthermore, the reduced distribution functions  $f_{\text{HCS}}$  and  $f_{0,\text{HCS}}$  have the scaling forms, inherited from Eq. (24),

$$\begin{aligned} f_{\text{HCS}}(\mathbf{v}_1, t) &= [\ell v(t)]^{-d} f_{\text{HCS}}^*[\mathbf{v}_1/v(t)], \\ f_{0,\text{HCS}}(\mathbf{v}_0, t) &= [\ell v(t)]^{-d} f_{0,\text{HCS}}^*[\mathbf{v}_0/v(t)]. \end{aligned} \quad (\text{B13})$$

Substitution of these expressions into the kinetic equations leads to

$$\begin{aligned} \frac{1}{2} \zeta^* \frac{\partial}{\partial \mathbf{v}_1^*} \cdot (\mathbf{v}_1 f_{\text{HCS}}^*) &= J_E^*[\mathbf{v}_1 | f_{\text{HCS}}^*], \\ \frac{1}{2} \zeta_0^* \frac{\partial}{\partial \mathbf{v}_0^*} \cdot (\mathbf{v}_1 f_{0,\text{HCS}}^*) &= J_{\text{EL}}^*[\mathbf{v}_0 | f_{0,\text{HCS}}^*, f_{\text{HCS}}^*], \end{aligned} \quad (\text{B14})$$

where the scaled position and velocity variables defined below Eq. (33) have been again introduced. The solution to these equations has been discussed elsewhere [26] and will not be considered further here.

Now consider more general spatially inhomogeneous states for the impurity particle, with the fluid still in the HCS. The probability density to find the impurity at a point  $\mathbf{q}_0$  in terms of the reduced distribution function is given by

$$P(\mathbf{q}_0, t) = \int d\mathbf{v}_0 f_0(\mathbf{q}_0, \mathbf{v}_0, t). \quad (\text{B15})$$

Integration of Eq. (B6) over the velocity  $\mathbf{v}_0$  gives the conservation law for probability

$$\partial_t P(\mathbf{q}_0, t) + \nabla_0 \cdot \mathbf{J}(\mathbf{q}_0, t) = 0, \quad (\text{B16})$$

$$\mathbf{J}(\mathbf{q}_0, t) = \int d\mathbf{v}_0 \mathbf{v}_0 f_0(\mathbf{q}_0, \mathbf{v}_0, t), \quad (\text{B17})$$

which, of course, are the same as Eqs. (57) and (58). The diffusion equation is obtained from a ‘‘normal’’ solution to the Boltzmann-Lorentz equation, where all space and time dependence occurs through  $P(\mathbf{q}_0, t)$  and  $T(t)$ . The linear relationship (B15) implies that such a solution has the form

$$f_0(\mathbf{q}_0, \mathbf{v}_0, t) = P(\mathbf{q}_0, t) h[\mathbf{v}_0/v(t)]. \quad (\text{B18})$$

The Chapman-Enskog method represents a normal solution to the kinetic equation as an expansion in the gradients,

$$f_0(\mathbf{q}_0, \mathbf{v}_0, t) = f_0^{(0)}(\mathbf{q}_0, \mathbf{v}_0, t) + \epsilon f_0^{(1)}(\mathbf{q}_0, \mathbf{v}_0, t) + \dots, \quad (\text{B19})$$

where  $\epsilon$  is a formal parameter representing the order of the spatial gradient. Similarly, the time derivative is obtained as an expansion in the gradients via the conservation equation,

$$\partial_t = \partial_t^{(0)} + \epsilon \partial_t^{(1)} + \dots \quad (\text{B20})$$

and the kinetic equation is written

$$(\partial_t + \epsilon \mathbf{v}_0 \cdot \nabla_0) f_0(\mathbf{q}_0, \mathbf{v}_0, t) = J_{\text{EL}}[\mathbf{q}_0, \mathbf{v}_0 | f_0(t), f_{\text{HCS}}(t)]. \quad (\text{B21})$$

Substitution of Eqs. (B19) and (B20) into Eq. (B21) gives to zeroth order in the spatial gradients

$$\partial_t^{(0)} f_0^{(0)} = J_{\text{EL}}[\mathbf{q}_0, \mathbf{v}_0 | f_0^{(0)}(t), f_{\text{HCS}}(t)], \quad (\text{B22})$$

which has the solution

$$f_0^{(0)}(\mathbf{q}_0, \mathbf{v}_0, t) = VP(\mathbf{q}_0, t) f_{\text{HCS}}(\mathbf{v}_0, t). \quad (\text{B23})$$

This gives  $\mathbf{J}^{(0)} = 0$  and, consequently,  $\partial_t^{(1)} P = 0$ . The first-order correction  $f_0^{(1)}$  is determined from

$$\partial_t^{(0)} f_0^{(1)} + (\partial_t^{(1)} + \mathbf{v}_0 \cdot \nabla_0) f_0^{(0)} = J_{\text{EL}}[\mathbf{q}_0, \mathbf{v}_0 | f_0^{(1)}(t), f_{\text{HCS}}(t)]. \quad (\text{B24})$$

The contribution from  $\partial_t^{(1)}$  vanishes, since it is proportional to  $\partial_t^{(1)} P(\mathbf{q}_0, t) = -\nabla_0 \cdot \mathbf{J}^{(0)}(\mathbf{q}_0, t) = 0$ . Also, the time derivative  $\partial_t^{(0)}$  can be expressed in terms of the cooling rate  $\zeta$ . Using once again the dimensionless variables defined in the main text, Eq. (B24) becomes

$$\mathcal{I}_{\text{EL}} f_0^{*(1)} - \frac{\zeta^*}{2} \frac{\partial}{\partial \mathbf{v}_0^*} \times (\mathbf{v}_0^* f_0^{*(1)}) = f_{0,\text{HCS}}^* \mathbf{v}_0^* \cdot \nabla_0^* P^*(\mathbf{q}_0^*, t), \quad (\text{B25})$$

where  $\mathcal{I}_{\text{EL}}$  is the linear Enskog-Lorentz collision operator,

$$\begin{aligned} \mathcal{I}_{\text{EL}} f_0^{*(1)} &\equiv \int d\mathbf{v}_1^* \int d\Omega \Theta(-\mathbf{g}_{01}^* \cdot \hat{\boldsymbol{\sigma}}) |\mathbf{g}_{01}^* \cdot \hat{\boldsymbol{\sigma}}| (\alpha_0^{-2} b_{01}^{-1} - 1) \\ &\quad \times f^*(\mathbf{v}_1^*) f_0^{*(1)}(\mathbf{q}_0^*, \mathbf{v}_0^*, t), \end{aligned} \quad (\text{B26})$$

that is the same as the collision operator  $\mathcal{I}$  in Eq. (99) if velocity correlations, associated to the two-particle distribution, are neglected in the latter. The solution to Eq. (B25) can be written in the form

$$f_0^{*(1)}(\mathbf{v}_0^*, \mathbf{q}^*, t) = \mathbf{X}(\mathbf{v}_0^*) \cdot \nabla_0^* P^*(\mathbf{q}^*, s), \quad (\text{B27})$$

where  $\mathbf{X}(\mathbf{v}_0^*)$  is the solution to the integral equation

$$\mathcal{I}_{\text{EL}} X_i(\mathbf{v}_0^*) - \frac{\zeta^*}{2} \frac{\partial}{\partial \mathbf{v}_0^*} \times [\mathbf{v}_0^* X_i(\mathbf{v}_0^*)] = f_{0,\text{HCS}}^*(v_0^*) v_{0i}^*. \quad (\text{B28})$$

Finally, the diffusion coefficient is identified from

$$\mathbf{J}^{(1)}(\mathbf{q}_0, t) = \int d\mathbf{v}_0 \mathbf{v}_0 f_0^{(1)}(\mathbf{q}_0, \mathbf{v}_0, t) = -D \nabla_0 P(\mathbf{q}_0, t) \quad (\text{B29})$$

to get

$$D^* = -\frac{2}{d\phi_{\text{HCS}}} \int d\mathbf{v}_0^* \mathbf{v}_0^* \cdot \mathbf{X}(\mathbf{v}_0^*). \quad (\text{B30})$$

These are the same results as those from Sec. IV, Eqs. (101) and (102).

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