

ON THE INTERMEDIATE TIME BEHAVIOR OF THE STRESS TENSOR AND VELOCITY AUTOCORRELATION FUNCTIONS FOR DENSE HARD SPHERE SYSTEMS

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It is conjectured that the large values at intermediate times of the stress tensor autocorrelation function, found in computer simulations, may be caused by a coupling of the stress tensor to pairs of slowly decaying extended heat modes of large wave number. Approximate expressions, amenable to numerical verification, are given for both the stress tensor and the velocity autocorrelation function.

Some time ago Erpenbeck and Wood [1] found in molecular dynamics calculations that for dense hard sphere systems the stress tensor autocorrelation function, occurring in the Green–Kubo formula for the shear viscosity, exhibits a slow decay of relatively large amplitude in the time regime between about 10 and 35 mean free times, which cannot be explained by standard mode-coupling theories [2]. These results were confirmed by Evans [3], who studied the frequency dependent viscosity $\eta(\omega)$ in non-equilibrium molecular dynamics simulations and found a behavior that could be fitted as $\eta(\omega) = \eta(0) + \alpha\omega^{1/2}$, again with a coefficient α much larger than could be explained by mode-coupling theory. Furthermore, Erpenbeck and Wood [1] also found that in the same density and time regime where the viscosity time correlation function exhibits the behavior mentioned above, the velocity autocorrelation function for a tagged hard sphere attains rather large negative values before returning to the (positive) long-time behaviour predicted by mode coupling theories.

In this paper I want to point out a possible cause for these phenomena and present some explicit formulae, the evaluation of which would provide a check on the validity of my conjecture.

De Schepper and Cohen [4,5] found in a study of the eigenfunctions and eigenvalues of the linearized Revised Enskog Equation that the hydrodynamic modes can be extended to extremely short wavelengths, that is wavelengths shorter than the hard sphere diameter. Even more remarkably, they found that at large densities one of the eigenvalues, namely that of the extended heat mode, exhibits a pronounced maximum close to zero (but negative) for a wave length, close to the hard sphere diameter, where the static structure factor is near a pronounced maximum. For example at the density of 0.625 times the close packing density, studied by Erpenbeck and Wood [1], the relaxation time of a heat mode at this maximum is about 20 mean free times. By comparing their predictions for the amplitude of the long time tails of the velocity autocorrelation function for two-dimensional hard disk systems to the results of computer simulations, Dorfman and Cohen could conclude, that within the time regime up till ~ 50 mean free times the hydrodynamic modes of the Enskog equation are a very good approximation to the exact hydrodynamic modes of hard sphere systems. De Schepper, Cohen and Zuilhof [5] provided strong evidence that this remains true for the extended hydrodynamic modes. Therefore it is natural to extend the standard mode coupling equations so as to include couplings to products of the extended modes. In particular products of heat modes with decay rates close to the minimal one may be expected to yield important contributions. In this vein De Schepper and Cohen studied the velocity autocorrelation function of a tagged particle [7]. They could show that the coupling of the tagged particle velocity to products of a diffusive mode for the

tagged particle motion (which is also slowly decaying at large densities) and a heat mode of opposite wave number for the fluid motion, yields the velocity autocorrelation function negative for intermediate times. They did have problems, however, reproducing quantitatively the molecular dynamics results of Erpenbeck and Wood.

It is my conjecture that in the case of the stress tensor autocorrelation function it is the coupling to pairs of heat modes with opposite wave number that is producing large contributions for intermediate times. The main cause for the large amplitude resulting from these contributions is the relatively large volume in wave number space from which contributing mode pairs can be chosen, namely a *spherical shell* of a width proportional to $t^{-1/2}$, where t is the time argument of the correlation function, about k_G , the wave number for which the decay rate is minimal. In contrast the mode pairs contributing to the standard $t^{-3/2}$ long time tails have wave numbers lying within a *sphere* of radius proportional to $t^{-1/2}$ around the origin.

Let us consider the correlation functions occurring in the Green–Kubo formula for the shear viscosity in some more detail. Following ref. [1] I write the Green–Kubo formula for the shear viscosity as the sum of three contributions

$$\eta = \eta^{kk} + 2\eta^{k\Phi} + \eta^{\Phi\Phi}, \quad (1)$$

with

$$\eta^{AB} = \lim_{t \rightarrow \infty} \eta^{AB}(t), \quad \eta^{AB}(t) = \beta \int_0^t dt' \rho_{\eta}^{AB}(t'), \quad \rho_{\eta}^{AB}(t) = \lim_{V \rightarrow \infty} V^{-1} \langle J_{\eta}^A(0) J_{\eta}^B(t) \rangle, \quad (2a-c)$$

where $\beta = 1/k_B T$, k_B is the Boltzmann constant and T is the temperature, the angular brackets denote an equilibrium average, V is the volume and $\lim_{V \rightarrow \infty}$ denotes the thermodynamic limit. For a periodic system of N identical particles of mass m , interacting through pair potentials $\varphi(\mathbf{r}_i - \mathbf{r}_j)$, the kinetic current J_{η}^k and the potential current J_{η}^{Φ} are generally given as tensorial components of the corresponding parts of the microscopic stress tensor,

$$J_{\eta}^k = T_{xy}^k, \quad J_{\eta}^{\Phi} = T_{xy}^{\Phi}, \quad \mathbf{T}^k = m \sum_i \mathbf{v}_i \mathbf{v}_i, \quad \mathbf{T}^{\Phi} = -\frac{1}{2} \sum_{i \neq j} \mathbf{r}_{ij} \partial \varphi(\mathbf{r}_{ij}) / \partial \mathbf{r}_{ij}, \quad (3a-d)$$

with $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$ and \mathbf{r}_i and \mathbf{v}_i denote the position and velocity of particle i . For hard spheres, due to the singular nature of the interparticle potential, (3d) is not well defined. Instead the potential part of the stress tensor is given as

$$\mathbf{T}^{\Phi} = m \sum_{\gamma=1}^{\infty} \mathbf{r}_{ij}(t_{\gamma}) \Delta \mathbf{v}_i(t_{\gamma}) \delta(t - t_{\gamma}), \quad (4)$$

where t_1, t_2, \dots denote the times at which binary collisions occur, i and j depend on γ and select the pair of spheres colliding at t_{γ} , $\mathbf{r}_{ij}(t_{\gamma})$ is the vector connecting their centers and $\Delta \mathbf{v}_i(t_{\gamma})$ is the change in velocity of particle i in this collision.

My starting point for calculating the time correlation functions ρ_{η}^{AB} in the time regime of interest is a linearized ring kinetic equation that under certain approximations can be obtained by the methods of [8]. Setting for the one-particle distribution function $f(\mathbf{x}, t) = n\varphi(\mathbf{v})\psi(\mathbf{x}, t)$, where $\mathbf{x} = (\mathbf{r}, \mathbf{v})$, n equals the equilibrium density and $\varphi(\mathbf{v})$ is the Maxwellian velocity distribution

$$\varphi(\mathbf{v}) = (\beta m / 2\pi)^{3/2} \exp(-\frac{1}{2} \beta m v^2), \quad (5)$$

the ring kinetic equation, in absence of an external potential, takes the form

$$(\partial / \partial t + \mathbf{v}_1 \cdot \partial / \partial \mathbf{r}_1) \psi(\mathbf{x}_1, t) = \Lambda_E(\mathbf{x}_1) \psi(\mathbf{x}_1, t) + \int_0^t d\tau R(\mathbf{x}_1, \tau) \psi(\mathbf{x}_1, t - \tau). \quad (6)$$

The linearized Enskog operator Λ_E is defined as

$$\Lambda_E(x_1) = n \int dx_2 \varphi(v_2) \bar{T}_{12} \left(g(\sigma)(1 + \mathcal{P}_{12}) + n \int dx_3 \varphi(v_3) H(\mathbf{r}_1, \mathbf{r}_2 | \mathbf{r}_3) \mathcal{P}_{13} \right), \quad (7)$$

where \mathcal{P}_{ij} is a permutation operator, exchanging the arguments x_i and x_j in its operand; $g(\sigma)$, with σ the hard sphere diameter, is the equilibrium pair correlation function for two spheres in contact and $H(\mathbf{r}_1, \mathbf{r}_2 | \mathbf{r}_3)$ is a three point position correlation function, given as the functional derivative [9]

$$H(\mathbf{r}_1, \mathbf{r}_2 | \mathbf{r}_3) = \delta g(\mathbf{r}_1, \mathbf{r}_2) / \delta n(\mathbf{r}_3), \quad (8)$$

where $g(\mathbf{r}_1, \mathbf{r}_2)$ is the pair correlation function, defined for general, non-uniform equilibrium states and (8) is evaluated for states of uniform density. Further the binary collision operator \bar{T}_{12} is given as

$$\bar{T}_{12} = |\mathbf{v}_{12} \cdot \hat{\mathbf{r}}_{12}| \delta(r_{12} - \sigma) [\theta(\mathbf{v}_{12} \cdot \hat{\mathbf{r}}_{12}) b_{\hat{\mathbf{r}}_{12}} - \theta(-\mathbf{v}_{12} \cdot \hat{\mathbf{r}}_{12})], \quad (9)$$

with $\mathbf{v}_{12} = \mathbf{v}_1 - \mathbf{v}_2$ and $\hat{\mathbf{r}}_{12} = \mathbf{r}_{12} / |\mathbf{r}_{12}|$. $\theta(x)$ is the Heaviside step function and $b_{\hat{\sigma}}$ is an operator changing \mathbf{v}_1 and \mathbf{v}_2 in its operand to \mathbf{v}'_1 and \mathbf{v}'_2 , defined as

$$\mathbf{v}'_1 = \mathbf{v}_1 - (\mathbf{v}_{12} \cdot \hat{\sigma}) \hat{\sigma}, \quad \mathbf{v}'_2 = \mathbf{v}_2 + (\mathbf{v}_{12} \cdot \hat{\sigma}) \hat{\sigma}. \quad (10)$$

The ring operator $R(x_1, \tau)$ is defined as

$$\begin{aligned} R(x_1, \tau) = & n \int dx_2 \varphi(v_2) \bar{T}_{12} \left(g(\sigma) + n \int dx_3 \varphi(v_3) H(\mathbf{r}_1, \mathbf{r}_2 | \mathbf{r}_3) (\mathcal{P}_{13} + \mathcal{P}_{23}) \right. \\ & \left. + \frac{1}{2} n^2 \int dx_3 \int dx_4 \varphi(v_3) \varphi(v_4) H(\mathbf{r}_1, \mathbf{r}_2 | \mathbf{r}_3, \mathbf{r}_4) \mathcal{P}_{13} \mathcal{P}_{24} \right) \\ & \times S_E(x_1, \tau) S_E(x_2, \tau) \left(g(\sigma) + n \int dx_5 \varphi(v_5) (\mathcal{P}_{15} + \mathcal{P}_{25}) H(\mathbf{r}_1, \mathbf{r}_2 | \mathbf{r}_5) \right. \\ & \left. + \frac{1}{2} n^2 \int dx_5 \int dx_6 \varphi(v_5) \varphi(v_6) \mathcal{P}_{15} \mathcal{P}_{26} H(\mathbf{r}_1, \mathbf{r}_2 | \mathbf{r}_5, \mathbf{r}_6) \right) T_{12} (1 + \mathcal{P}_{12}). \end{aligned} \quad (11)$$

Here $H(\mathbf{r}_1, \mathbf{r}_2 | \mathbf{r}_3, \mathbf{r}_4)$ is a four point correlation function, defined as

$$H(\mathbf{r}_1, \mathbf{r}_2 | \mathbf{r}_3, \mathbf{r}_4) = \delta^2 g(\mathbf{r}_1, \mathbf{r}_2) / \delta n(\mathbf{r}_3) \delta n(\mathbf{r}_4). \quad (12)$$

The binary collision operator T_{12} is defined similarly to \bar{T}_{12} as

$$T_{12} = |\mathbf{v}_{12} \cdot \hat{\mathbf{r}}_{12}| \delta(r_{12} - \sigma) \theta(\mathbf{v}_{12} \cdot \hat{\mathbf{r}}_{12}) (b_{\hat{\mathbf{r}}_{12}} - 1), \quad (13)$$

and the Enskog propagator is defined as

$$S_E(x_1, t) = \exp[\Lambda_E(x_1)t] \left(1 + n \int dx_2 \varphi(v_2) h(\mathbf{r}_1, \mathbf{r}_2) \mathcal{P}_{12} \right), \quad (14)$$

with $h(\mathbf{r}_1, \mathbf{r}_2) = g(\mathbf{r}_1, \mathbf{r}_2) - 1$.

The correlation functions at intermediate times that are of interest to us, are obtained by applying (6) to an initial distribution function of the form $\psi(x_1) = (\mathbf{v}_1 \cdot \hat{\mathbf{I}}) \exp(\mathbf{i}\mathbf{k} \cdot \mathbf{r}_1)$, with $\hat{\mathbf{I}}$ a fixed unit vector satisfying $(\hat{\mathbf{I}} \cdot \mathbf{k}) = 0$, and considering the limit $k \rightarrow 0$. In doing so one has to make some approximations; the most important ones are based on the facts that (i) the short time behavior is dominated by the Enskog operator, and (ii) the intermediate time behavior is governed by terms containing just one ring operator. Then the following expressions for the correlation functions in the time range of interest can be extracted:

$$\rho_n^{AB}(t) = \frac{1}{2\beta^2 (2\pi)^3} \int d\mathbf{q} \sum_{\alpha} \sum_{\beta} \exp\{[z_{\alpha}(q) + z_{\beta}(q)]t\} V_{\alpha\beta}^A(\mathbf{q}) V_{\alpha\beta}^B(-\mathbf{q}), \quad (15)$$

with vertex functions $V_{\alpha\beta}$ defined as

$$\begin{aligned}
V_{\alpha\beta}^k(\mathbf{q}) &= n\sigma^2 \int d\mathbf{v}_1 \int d\mathbf{v}_2 \varphi(v_1)\varphi(v_2) \int d\hat{\boldsymbol{\sigma}} \theta(\mathbf{v}_{12} \cdot \hat{\boldsymbol{\sigma}}) (\mathbf{v}_{12} \cdot \hat{\boldsymbol{\sigma}}) (b_{\hat{\boldsymbol{\sigma}}} - 1) \\
&\times \{ (1 + \mathcal{P}_{12}) [-\Lambda_E(x_1)]^{-1} \beta m (\mathbf{v}_1 \cdot \hat{\boldsymbol{k}}) (\mathbf{v}_1 \cdot \hat{\mathbf{1}}) \} \{ [\exp(i\mathbf{q} \cdot \hat{\boldsymbol{\sigma}}) g(\sigma) + \exp(\frac{1}{2} i\mathbf{q} \cdot \hat{\boldsymbol{\sigma}}) H(\mathbf{q}, \hat{\boldsymbol{\sigma}})] \\
&\times \{ P(1)[1 - P(2)] + [1 - P(1)]P(2) \} + \exp(i\mathbf{q} \cdot \hat{\boldsymbol{\sigma}}) g(\sigma) [1 - P(1)] [1 - P(2)] \} \Psi_{\alpha}(-\mathbf{q}, \mathbf{v}_1) \Psi_{\beta}(\mathbf{q}, \mathbf{v}_2), \quad (16a)
\end{aligned}$$

$$\begin{aligned}
V_{\alpha\beta}^{\Phi}(\mathbf{q}) &= \int d\mathbf{v}_1 \int d\mathbf{v}_2 \varphi(v_1)\varphi(v_2) \left(-(\hat{\boldsymbol{k}} \cdot \hat{\mathbf{q}}) (\hat{\mathbf{1}} \cdot \hat{\mathbf{q}}) q \frac{dnC(q)}{dq} + n\sigma^3 \beta m \int d\hat{\boldsymbol{\sigma}} \theta(\mathbf{v}_{12} \cdot \hat{\boldsymbol{\sigma}}) (\mathbf{v}_{12} \cdot \hat{\boldsymbol{\sigma}})^2 (\hat{\boldsymbol{k}} \cdot \hat{\boldsymbol{\sigma}}) (\hat{\mathbf{1}} \cdot \hat{\boldsymbol{\sigma}}) \right. \\
&\times [\exp(i\mathbf{q} \cdot \hat{\boldsymbol{\sigma}}) g(\sigma) + \exp(\frac{1}{2} i\mathbf{q} \cdot \hat{\boldsymbol{\sigma}}) H(\mathbf{q}, \hat{\boldsymbol{\sigma}})] \{ P(1)[1 - P(2)] + [1 - P(1)]P(2) \} \\
&\left. + \exp(i\mathbf{q} \cdot \hat{\boldsymbol{\sigma}}) g(\sigma) [1 - P(1)] [1 - P(2)] \right) \Psi_{\alpha}(-\mathbf{q}, \mathbf{v}_1) \Psi_{\beta}(\mathbf{q}, \mathbf{v}_2) + \frac{4}{15} \pi n \sigma^3 g(\sigma) V_{\alpha\beta}^k(\mathbf{q}). \quad (16b)
\end{aligned}$$

In these equations $\hat{\boldsymbol{\sigma}}$ is a unit vector and $\int d\hat{\boldsymbol{\sigma}}$ runs over the unit sphere. The inverse Enskog operator acts within the space of functions orthogonal to its zero eigenfunctions under the scalar product

$$(\Psi|\chi) = \int d\mathbf{v} \varphi(v) \Psi^*(\mathbf{v}) \chi(\mathbf{v}). \quad (17)$$

The function $H(\mathbf{q}, \hat{\boldsymbol{\sigma}})$ is defined as

$$H(\mathbf{q}, \hat{\boldsymbol{\sigma}}) = \int d\mathbf{r} \exp(i\mathbf{q} \cdot \mathbf{r}) H(-\frac{1}{2} \hat{\boldsymbol{\sigma}} \sigma, \frac{1}{2} \hat{\boldsymbol{\sigma}} \sigma | \mathbf{r}), \quad (18)$$

and $C(q)$ is the Fourier transform of the direct correlation function, satisfying $nC(q) = -1/S(q)$ with $S(q)$ the static structure factor,

$$S(q) = \int d\mathbf{r} \exp(i\mathbf{q} \cdot \mathbf{r}) [\delta(\mathbf{r}) + nh(r)]. \quad (19)$$

The operator $P(i)$ is defined as

$$P(i) = \int d\mathbf{v}_i \varphi(v_i). \quad (20)$$

It projects functions of \mathbf{v}_i upon the unit function. The sums over α and β run over the eigenvalues $z_{\alpha}(q)$ of the operator $-i\mathbf{q} \cdot \mathbf{v} + \Lambda_E(\mathbf{q}, \mathbf{v})$, with $\Lambda_E(\mathbf{q}, \mathbf{v})$ the spatial Fourier transform of $\Lambda_E(x)$. The corresponding right and left eigenfunctions of this operator are $\Psi_{\alpha}(\mathbf{q}, \mathbf{v})$ and $\Phi_{\alpha}(\mathbf{q}, \mathbf{v})$ respectively, chosen so as to satisfy the relations [4]

$$\Psi_{\alpha}(\mathbf{q}, \mathbf{v}_1) = \{1 + [S(q) - 1]P(1)\} \Phi_{\alpha}^*(\mathbf{q}, \mathbf{v}_1), \quad (\Phi_{\alpha}(\mathbf{q}, \mathbf{v}_1) | \Psi_{\alpha}(\mathbf{q}, \mathbf{v}_1)) = 1. \quad (21a,b)$$

By the expansion of the Enskog propagators within the ring operator in terms of eigenfunctions, the contributions of pairs of slowly decaying heat modes are exposed explicitly. Preliminary calculations [10] of (15) indicate a $\rho_{\eta}^{\Phi\Phi}$ in the same order of magnitude as found in the computer simulations of Erpenbeck and Wood [1]. However these calculations are very delicate, for instance the heat mode for q near q_G , where the decay rate is minimal, is close to the unit function [4], hence the first term on the right-hand side of (16b) might be expected to be dominant, but its coefficient $q dC/dq$ becomes zero at a value of q very close to q_G . Furthermore, assuming ρ_{η}^{kk} to be insignificant compared to $\rho_{\eta}^{\Phi k}$, as seems to be indicated by the results of ref. [1], one sees from (15) and (16) that $\rho_{\eta}^{\Phi\Phi}$ contains a contribution $\approx \frac{8}{15} \pi n \sigma^3 g(\sigma) \rho_{\eta}^{\Phi k}$, which according to ref. [1] at a density of 0.625 times the close packing density would amount to approximately $-0.3 \rho_{\eta}^{\Phi\Phi}$. Hence we must conclude that for a reliable calculation of the viscosity time correlation functions ρ_{η}^{AB} a precise knowledge of the heat mode eigenfunction and eigenvalue are required, as well as a fairly good knowledge of the function $H(\mathbf{q}, \hat{\boldsymbol{\sigma}})$.

By completely similar calculations one may obtain an expression for the intermediate time behavior of the tagged particle velocity autocorrelation function, of the form

$$\rho_D(t) = \langle \mathbf{v}_1(0) \cdot \mathbf{v}_1(t) \rangle \approx \frac{1}{n(2\pi)^3} \int d\mathbf{q} \sum_{\alpha} \sum_{\beta} \exp \{ [z_{\alpha}^s(\mathbf{q}) + z_{\beta}^s(\mathbf{q})] t \} |v_{\alpha\beta}^D(\mathbf{q})|^2, \quad (22)$$

with

$$v_{\alpha\beta}^D(\mathbf{q}) = n\sigma^2 \int d\mathbf{v}_1 \int d\mathbf{v}_2 \varphi(v_1)\varphi(v_2) \int d\hat{\sigma} \theta(\mathbf{v}_{12} \cdot \hat{\sigma})(\mathbf{v}_{12} \cdot \hat{\sigma})(b_{\hat{\sigma}} - 1) [-\Lambda_E^s(x_1)]^{-1} \\ \times \mathbf{v}_1 \{ \exp(i\mathbf{q} \cdot \hat{\sigma}\sigma)g(\sigma) + \exp(\frac{1}{2}i\mathbf{q} \cdot \hat{\sigma}\sigma)H(\mathbf{q}, \hat{\sigma})P(2) \} \Psi_{\alpha}^s(-\mathbf{q}, \mathbf{v}_1) \Psi_{\beta}^s(\mathbf{q}, \mathbf{v}_2). \quad (23)$$

Here, Λ_E^s is the Lorentz-Enskog operator defined as

$$\Lambda_E^s = ng(\sigma) \int dx_2 \varphi(v_2) \bar{T}_{12}, \quad (24)$$

and z_{α}^s runs over the eigenvalues of the operator $-i\mathbf{k} \cdot \mathbf{v} + \Lambda_E^s$. The corresponding right and left eigenfunctions, Ψ_{α}^s and Φ_{α}^s respectively, satisfy the relations [4] $\Phi_{\alpha}^s(\mathbf{q}, \mathbf{v}) = \Psi_{\alpha}^{s*}(\mathbf{q}, \mathbf{v})$ and $(\Phi_{\alpha}^s(\mathbf{q}, \mathbf{v}) | \Psi_{\alpha}^s(\mathbf{q}, \mathbf{v})) = 1$.

The calculations of De Schepper and Cohen [7] were based on an expression for $v_{\alpha\beta}^D(\mathbf{q})$ in (22) different from that given by (23). The disagreement between theory and simulation results found by these authors might well be due to this difference. Preliminary calculations [10] of (22), using (23) for $v_{\alpha\beta}^D$, indicate a fair agreement with simulation results, but a more precise knowledge of eigenfunctions and eigenvalues is needed again for a definite judgement of the merits of these equations.

A detailed derivation of the results presented here will appear elsewhere.

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