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Long time behavior of the Burnett transport coefficients

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The long time behavior of the integrands in the expressions for the Burnett transport coefficients for a system of hard spheres or disks is studied using kinetic theory. It is shown that, in the ring approximation, the kinetic parts have hydrodynamic tails behaving as $t^{1-d/2}$. It follows that the expressions for the Burnett transport coefficients are all divergent if this behavior persists for $t\to\infty$.

I. INTRODUCTION

The generalization of the hydrodynamic equations beyond the Navier–Stokes approximation is a subject which has received a great deal of attention over the last decades.[1] In particular, the structure of the so-called Burnett hydrodynamic equations has been derived starting from the Liouville equation and expressions for the associated transport coefficients in terms of time correlation functions have been given.[2,3] The Burnett equations correspond to the next order correction to the Navier–Stokes equations if one assumes that the transport equations can be expanded in gradients of the hydrodynamic variables. Nevertheless, theoretical arguments as well as computer studies have been accumulating evidence that the hydrodynamic fluxes are not analytic functions of the gradients present in the system.[4] The origin of this behavior seems to be closely related to the hydrodynamic long time tails. In fact, it has been shown that for two-dimensional systems there is a time interval over which the correlation functions appearing in the expressions of the Navier–Stokes transport coefficients have a t^{-1} decay. If this behavior persists for $t\to\infty$, the corresponding transport coefficients are divergent. In the case of three-dimensional systems, the decay in time of the correlation functions is as $t^{-3/2}$ and it is consistent with the existence of the Navier–Stokes transport coefficients in three dimensions.

The purpose of this paper is to study the transport coefficients associated with the higher gradient corrections to the Navier–Stokes equations. Although, explicit calculations will be restricted to Burnett order, for which expressions of the transport coefficients are known, some general conclusions will be presented here, and developed in a more detailed way in a later paper. We will use kinetic theory for a moderately dense gas of hard spheres, and our procedure will be essentially the one already used by several authors to study the long time behavior of the correlation functions associated with the Navier–Stokes transport coefficients.[5] Then, we show that the expressions for both the linear and nonlinear Burnett transport coefficients appear to be divergent for two- and three-dimensional systems. More previsely, they are expressed as time integrals extended up to infinity of functions decaying in time as $t^{1-d/2}$, where d is the dimensionality of the system.

The results presented in this paper are consistent with recent studies on systems having a steady-state shear flow.[6] Nevertheless, it must be pointed out that there is no Burnett order in such a problem. More closely related calculations have been carried out by Ernst and Dorfman.[7] These authors have shown that the hydrodynamic frequencies do not have analytic expansions in powers of the wave number. As a consequence, the linearized Burnett equations can not properly describe the hydrodynamic behavior of a fluid. Then, using mode coupling theory, they show that the linear Burnett transport coefficients are divergent. Because of the Fourier representation they introduce, their transport coefficients are linear combinations of the (linear) Burnett coefficients considered in this paper. In the low density limit their results agree with the values obtained here from kinetic theory, when the appropriated combinations are considered.

As said above, two general theories for transport equations to Burnett order have been worked out in Refs. 2 and 3. The general results of both theories are equivalent. The specialization to the case of a simple fluid has also been considered,[2,8] but some errors occur in these calculations. For this reason, and also because they are the starting point of our study, we give in the Appendix A corrected expressions for the complete set of Burnett coefficients.

The plan of the paper is as follows. In Sec. II we introduce distribution functions that are suitable for the calculation of the linear Burnett transport coefficients. These functions obey a hierarchy of equations that is truncated by using the ring approximation. In Sec. III the formal solution of the kinetic equation is used to compute the long time behavior of the correlation functions appearing in the expressions of the linear Burnett coefficients. It is shown that they all diverge in two and three dimensions. A comparison of our results with corresponding results by Ernst and Dorfman is made in Appendix B. In Sec. IV the above theory is extended to include correlation functions involving three dynamical variables. In the low density limit our results agree with those first derived by Dufty, as shown in Appendix C. In the ring approximation, the calculation of the long time behavior reduces to the problem already solved in Sec. III and the expressions of the nonlinear coefficients diverge in the same way as those of the linear ones. As an example we consider the long time limit of the viscometric functions to Burnett order.

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II. TWO-POINT CORRELATION FUNCTIONS. RING APPROXIMATION

Let us consider the linear approximation of the hydrodynamic equations to Burnett order. If we neglect terms containing the product of the gradients of two thermodynamical variables, Eqs. (A1) and (A2) become

$$\langle\tau_{ij}(\mathbf{r})\rangle_t^{(D)} = -\eta(\mathbf{r},t)\left[\frac{\partial u_i(\mathbf{r},t)}{\partial r_j} + \frac{\partial u_j(\mathbf{r},t)}{\partial r_i} - \frac{2}{d}\delta_{ij}\nabla\cdot\mathbf{u}(\mathbf{r},t)\right]$$
$$-\delta_{ij}\zeta(\mathbf{r},t)\nabla\cdot\mathbf{u}(\mathbf{r},t) - \frac{2\eta_1(\mathbf{r},t)}{mn(\mathbf{r},t)}\frac{\partial^2 p(\mathbf{r},t)}{\partial r_i\partial r_j}$$
$$+\delta_{ij}\eta_4(\mathbf{r},t)\nabla^2 T(\mathbf{r},t) - \delta_{ij}\frac{\eta_2(\mathbf{r},t)}{mn(\mathbf{r},t)}\nabla^2 p(\mathbf{r},t)$$
$$+\eta_3(\mathbf{r},t)\frac{\partial^2 T(\mathbf{r},t)}{\partial r_i\partial r_j} \tag{1}$$

and

$$\langle J_i^E(\mathbf{r})\rangle_t^{(D)} = -\lambda(\mathbf{r},t)\frac{\partial}{\partial r_i}T(\mathbf{r},t)$$
$$+\left[\lambda_2(\mathbf{r},t) - \lambda_1(\mathbf{r},t)T(\mathbf{r},t)\left(\frac{\partial p(\mathbf{r},t)}{\partial e(\mathbf{r},t)}\right)_n\right]$$
$$\times\frac{\partial}{\partial r_i}\nabla\cdot\mathbf{u}(\mathbf{r},t) + \lambda_3(\mathbf{r},t)\nabla^2 u_i(\mathbf{r},t) \ . \tag{2}$$

Here η, ζ, and λ are the usual (Navier–Stokes) shear viscosity, bulk viscosity, and thermal conductivity, respectively, and are given by the Green–Kubo formula. The other transport coefficients η_1, η_2, η_3, η_4, λ_1, λ_2, and λ_3 are the linear Burnett transport coefficients. Their expressions are given in the Appendix A, where the notation is also explained.

We are going to consider the calculation of the long time behavior of the correlation functions appropriate to the linear Burnett transport coefficients for a system of hard spheres or hard disks. In fact, the theory will be formulated in such a general way that it will also include all the Navier–Stokes transport coefficients and some of the nonlinear Burnett ones. In the range of approximation we are going to consider we can confine our attention to the kinetic parts of the dynamical variables appearing in the correlation functions. The physical idea is the following. The collisional or potential transfer of momentum and energy is due to interactions between particles that are on opposite sides of an imaginary surface. Of course, this contribution vanishes when the difference in position of colliding particles is neglected, as it is the case in the low-density approximation we will consider here. An explicit calculation of the potential contributions to the correlation function expressions of the transport coefficients confirms the above ideas.

The kinetic part of the fluxes, and also of the conserved quantities, are expressed as sums of one-particle variables. Then, it is easily seen that the correlation functions we want to study have the general structure

$$\rho(\tau) = \langle F(\tau)F'(0)\rangle \ , \tag{3}$$

where the angular brackets denote equilibrium averages and

$$F = \sum_{\alpha=1}^{N} F_1(x_\alpha) \ ,$$
$$F' = \sum_{\alpha=1}^{N} F_1'(x_\alpha) \ ,$$
$$F(\tau) = e^{\tau L}F \ . \tag{4}$$

Here $x_\alpha \equiv \{\mathbf{r}_\alpha, \mathbf{p}_\alpha\}$ and L is the Liouville operator of the system.

We will assume without loss of generality that

$$\int dx\, F_1(x) f_{1,\mathrm{eq}}(x) = \int dx\, F_1'(x) f_{1,\mathrm{eq}}(x) = 0 \ , \tag{5}$$

where $f_{1,\mathrm{eq}}(x)$ is the one particle equilibrium distribution function. In order to study Eq. (3) it is convenient to introduce a distribution function $K_1(x_1,\ldots,x_N;\tau/z)$ defined by

$$K_1(x_1,\ldots,x_N;\tau/z)$$
$$= \exp[-\tau L(x)]\left[\sum_{\alpha=1}^{N}\delta(z-x_\alpha)f_{\mathrm{eq}}(x_1,\ldots,x_N)\right]$$
$$= \sum_\alpha \delta[z - x_\alpha(-\tau)]f_{\mathrm{eq}}(x_1,\ldots,x_N) \ , \tag{6}$$

where $f_{\mathrm{eq}}(x_1,\ldots,x_N)$ is the equilibrium distribution of the system. The function K_1 obeys the Liouville equation

$$\frac{\partial}{\partial\tau}K_1(x_1,\ldots,x_N;\tau/z) = -L(x)K_1(x_1,\ldots,x_N;\tau/z). \tag{7}$$

From Eq. (6) we define a set of reduced distributions as

$$f_{s/1}(y_1,\ldots,y_s;\tau/z)$$
$$= \sum_{\substack{\alpha_1 \\ \alpha_1\neq\cdots\neq\alpha_s}}\cdots\sum_{\alpha_s}\int dx_1\ldots dx_N\,\delta(y_1 - x_{\alpha_1})\ldots\delta(y_s - x_{\alpha_s})$$
$$\times K_1(x_1,\ldots,x_N;\tau/z) \ . \tag{8}$$

In particular, it is

$$f_{1/1}(y_1;\tau/z) = \int dx_1\ldots dx_N \sum_{\alpha_1}\delta[y_1 - x_{\alpha_1}(\tau)]$$
$$\times\sum_{\alpha_2}\delta(z - x_{\alpha_2})f_{\mathrm{eq}}(x_1,\ldots,x_N) \tag{9}$$

and this is the function that is required in order to evaluate the correlation function (3). We have

$$\rho(\tau) = \int dy\int dz\, F_1(y)F_1(z)f_{1/1}(y;\tau/z) \ . \tag{10}$$

From the Liouville equation (7) we can derive by direct integration a hierarchy of equations:

$$\partial_\tau f_{1/1}(y_1;\tau/z) = -L_1^0 f_{1/1}(y_1;\tau/z)$$
$$-\int dy_2\, L_{12}' f_{2/1}(y_1,y_2;\tau/z) \ , \tag{11a}$$

$$\partial_\tau f_{2/1}(y_1,y_2;\tau/z) = -(L_1^0 + L_2^0)f_{2/1}(y_1,y_2;\tau/z)$$
$$-L_{12}' f_{2/1}(y_1,y_2;\tau/z) - \int dy_3\,(L_{13}' + L_{23}')f_{3/1}(y_1,y_2,y_3;\tau/z), \tag{11b}$$

etc. Here it is

$$L_\alpha^0 = \mathbf{v}_\alpha \cdot \frac{\partial}{\partial \mathbf{r}_\alpha} \tag{12}$$

and

$$L'_{\alpha\beta} = -\frac{1}{2}\frac{\partial V(r_{\alpha\beta})}{\partial \mathbf{r}_{\alpha\beta}} \cdot \left(\frac{\partial}{\partial \mathbf{p}_\alpha} - \frac{\partial}{\partial \mathbf{p}_\beta}\right) . \tag{13}$$

These operators only act on the y's variables.

Hierarchies very similar, and even equivalent, to the one defined by Eqs. (11) have been considered by many authors. A very enlightening review has been carried out by Ernst and Cohen.[9] We have written the equations for a smooth potential, but in the following we will consider a system of hard spheres or disks for which calculations are much simpler. For this case, Eqs. (11) are still formally valid if we replace the operator $L'_{\alpha\beta}$ with[9]

$$L'_{\alpha\beta} = -\overline{T}_-(\alpha\beta) , \tag{14}$$

where $\overline{T}_-$ is the binary collision operator

$$\overline{T}_-(\alpha\beta) = \sigma^{d-1}\int_{\mathbf{v}_{\alpha\beta}\cdot\boldsymbol{\sigma}>0} d\hat{\sigma}\, |\mathbf{v}_{\alpha\beta}\cdot\hat{\sigma}|\,[\delta(\mathbf{r}_{\alpha\beta}-\boldsymbol{\sigma})b_\sigma - \delta(\mathbf{r}_{\alpha\beta}+\boldsymbol{\sigma})] . \tag{15}$$

In this expression, d is the dimension of the system, σ is the diameter of the hard spheres, $\hat{\sigma}$ a unit vector in the direction of the line of centers of the colliding pair, and $\boldsymbol{\sigma} = \sigma\hat{\sigma}$. The operator b_σ changes the velocities $\mathbf{v}_\alpha$ and $\mathbf{v}_\beta$ of the two particles before the collision by the velocities after the collision $\mathbf{v}_\alpha^*$, $\mathbf{v}_\beta^*$.

Next, we introduce the cluster functions $\chi_{s/1}$ defined by

$$f_{1/1}(y_1; \tau/z) = f_{1,\mathrm{eq}}(y_1)f_{1,\mathrm{eq}}(z) + \chi_{1/1}(y_1; \tau/z) , \tag{16a}$$

$$\begin{aligned} f_{2/1}(y_1, y_2; \tau/z) &= f_{1,\mathrm{eq}}(y_1)f_{1,\mathrm{eq}}(y_2)f_{1,\mathrm{eq}}(z) \\ &\quad + f_{1,\mathrm{eq}}(y_1)\chi_{1/1}(y_2; \tau/z) + f_{1,\mathrm{eq}}(y_2)\chi_{1/1}(y_1; \tau/z) \\ &\quad + f_{1,\mathrm{eq}}(z)g_{2,\mathrm{eq}}(y_1, y_2) + \chi_{2/1}(y_1, y_2; \tau/z) , \end{aligned} \tag{16b}$$

etc. This is the adaptation to our problem of the definition of the Ursell functions in equilibrium statistical mechanics and involves the equilibrium correlation functions $g_{s,\mathrm{eq}}(x_1, \ldots, x_s)$. The cluster functions also satisfy a set of hierarchy equations that follows directly from Eqs. (11) and (16):

$$[\partial_\tau + L_1^0 - \lambda(1)]\chi_{1/1}(y_1; \tau/z) = \int dy_2\, \overline{T}_-(12)\chi_{2/1}(y_1, y_2; \tau/z) , \tag{17a}$$

$$\begin{aligned} [\partial_\tau + L_1^0 + L_2^0 - \overline{T}_-(12) - \Lambda(12)]\chi_{2/1}(y_1, y_2; \tau/z) &= \overline{T}_-(12)(1+P_{12})\chi_{1/1}(y_1; \tau/z)f_{1,\mathrm{eq}}(y_2) \\ &\quad + (1+P_{12})\int dy_3\, \overline{T}_-(13)[(1+P_{13})g_{2,\mathrm{eq}}(y_1, y_2)\chi_{1/1}(y_3; \tau/z) + \chi_{3/1}(y_1, y_2, y_3; \tau/z)] , \end{aligned} \tag{17b}$$

etc. The operator $P_{\alpha\beta}$ permutes the particle labels (α, β), $\lambda(1)$ is the linearized Boltzmann collision operator for hard disks or hard spheres defined by

$$\lambda(1) = \int dy_2\, \overline{T}_-(12)(1+P_{12})f_{1,\mathrm{eq}}(y_2) \tag{18}$$

and

$$\Lambda(12) = \lambda(1) + \lambda(2) . \tag{19}$$

In order to analyze and solve the hierarchy (17) it is convenient to introduce the Laplace transform

$$\tilde{F}(\epsilon) = \int_0^\infty dt\, e^{-\epsilon t} F(t) , \tag{20}$$

and write

$$[\epsilon + L_1^0 - \lambda(1)]\tilde{\chi}_{1/1}(y_1; \epsilon/z) = \chi_{1/1}(y_1; \tau=0/z) + \int dy_2\, \overline{T}_-(12)\tilde{\chi}_{2/1}(y_1, y_2; \epsilon/z) , \tag{21a}$$

$$\begin{aligned} [\epsilon + L_1^0 + L_2^0 - \overline{T}_-(12) - \Lambda(12)]\tilde{\chi}_{2/1}(y_1, y_2; \epsilon/z) &= \overline{T}_-(12)(1+P_{12})\tilde{\chi}_{1/1}(y_1; \epsilon/z)f_{1,\mathrm{eq}}(y_2) \\ &\quad + (1+P_{12})\int dy_3\, \overline{T}_-(13)[(1+P_{13})g_{2,\mathrm{eq}}(y_1, y_2)\tilde{\chi}_{1/1}(y_3; \epsilon/z) \\ &\quad + \tilde{\chi}_{3/1}(y_1, y_2, y_3; \epsilon/z)] + \chi_{2/1}(y_1, y_2; \tau=0/z) . \end{aligned} \tag{21b}$$

These equations will be solved in the so-called ring approximation.[10] The idea is to take into account the contributions of uncorrelated binary collision events, given by the Boltzmann term, and also of those events giving the most divergent contributions in each order in the density expansion of the correction to the Boltzmann equation, the ring events. These are expected to give the dominant contributions in each order of the density after several mean free times and for sufficiently low densities. The ring events are collected and summed together leading to a resummed collision operator. The results obtained in this way are only valid for moderately dense gases, they represent the first order correction in density to the Boltzmann equation.

The initial values in Eqs. (21) are given by

$$\chi_{1/1}(y_1; \tau=0/z) = \delta(y_1 - z)f_{1,\mathrm{eq}}(y_1) + g_{2,\mathrm{eq}}(y_1, z) \tag{22}$$

and

$$\chi_{2/1}(y_1, y_2; \tau=0/z) = [\delta(y_1-z) + \delta(y_2-z)]g_{2,\mathrm{eq}}(y_1, y_2) + g_{3,\mathrm{eq}}(y_1, y_2, z) . \tag{23}$$

These expressions are obtained by comparing the definition (8) of $f_{s/1}$ and the cluster expansion (16) for $\tau=0$. When one starts from a set of BBGKY equations like Eq. (21) the ring approximation is easily introduced by making the following simplifications.[5,11]

(i) We neglect in Eq. (21b) the term $\tilde{\chi}_{3/1}(y_1, y_2, y_3; \epsilon/z)$. In this way, Eqs. (21a) and (21b) define a closed equation. To analyze the contributions associated with this term, one has to consider the third equation of the hierarchy. The analysis shows that the neglected events are less divergent than the ring events of the same order in the density.

(ii) In addition, we will neglect the contributions in Eq. (21b) arising from the initial condition term $\chi_{2/1}(y_1, y_2; \tau = 0/z)$ because they are also less divergent and of higher order in density than the dominant term. This is a consequence of the fact that $g_{2,\mathrm{eq}}(y_1, y_2)$ is one order higher in the density than $f_{1,\mathrm{eq}}(y_1) f_{1,\mathrm{eq}}(y_2)$. Of course, a similar argument holds for $g_{3,\mathrm{eq}}(y_1, y_2, y_3)$.

(iii) The term proportional to

$$\int dy_3\, \overline{T}_-(13)[(1+P_{13}) g_{2,\mathrm{eq}}(y_1, y_2) \tilde{\chi}_{1/1}(y_3; \epsilon/z)]$$

is also of higher order in density than the leading term, due to the $\overline{T}_-(13)$ operator that is nonzero only if the particles 1 and 3 are within a distance σ. Thus, it will not be considered.

With these approximations Eqs. (21) lead to the closed equation

$$\tilde{\chi}_{1/1}(y_1; \epsilon/z) = [\epsilon + L_1^0 - \lambda(1) - \tilde{M}(1; \epsilon)]^{-1} \times [g_{2,\mathrm{eq}}(y_1, z) + \delta(y_1 - z) f_{1,\mathrm{eq}}(y_1)], \tag{24}$$

where $\tilde{M}$ is defined by

$$\tilde{M}(1; \epsilon) = \int dy_2\, \overline{T}_-(12)[\epsilon + L_1^0 + L_2^0 - \overline{T}_-(12) - \wedge(12)]^{-1} \times \overline{T}_-(12)(1 + P_{12}) f_{1,\mathrm{eq}}(y_2). \tag{25}$$

Nevertheless, this expression still retains contributions of different orders. Using the identity between operators $A^{-1} = B^{-1} + B^{-1}(B - A)A^{-1}$ we can write

$$[\epsilon + L_1^0 - \lambda(1) - \tilde{M}(1; \epsilon)]^{-1} = [\epsilon + L_1^0 - \lambda(1)]^{-1} \sum_{p=0}^{\infty} \{\tilde{M}(1; \epsilon)[\epsilon + L_1^0 - \lambda(1)]^{-1}\}^p \tag{26}$$

and

$$\tilde{M}(1; \epsilon) = \int dy_2\, \overline{T}_-(12)[\epsilon + L_1^0 + L_2^0 - \wedge(12)]^{-1} \times \sum_{q=0}^{\infty} \{\overline{T}_-(12)[\epsilon + L_1^0 + L_2^0 - \wedge(12)]^{-1}\}^q \times \overline{T}_-(12)(1 + P_{12}) f_{1,\mathrm{eq}}(y_2). \tag{27}$$

(iv) When the expansions (26) and (27) are analyzed it is seen that in the ring approximation we must take in Eq. (24),

$$[\epsilon + L_1^0 - \lambda(1) - \tilde{M}(1; \epsilon)]^{-1} = [\epsilon + L_1^0 - \lambda(1)]^{-1} + [\epsilon + L_1^0 - \lambda(1)]^{-1} \tilde{R}(1; \epsilon)[\epsilon + L_1^0 - \lambda(1)]^{-1}, \tag{28}$$

where $\tilde{R}(1; \epsilon)$ is the so-called ring collision operator

$$\tilde{R}(1; \epsilon) = \int dy_2\, \overline{T}_-(12)[\epsilon + L_1^0 + L_2^0 - \wedge(12)]^{-1} \times \overline{T}_-(12)(1 + P_{12}) f_{1,\mathrm{eq}}(y_2). \tag{29}$$

In particular, we eliminate in this way repeated ring contributions.

(v) Also, the term containing $g_{2,\mathrm{eq}}(y_1, z)$ can be neglected in Eq. (24) because, as we said above, it is of higher order in density.

So, in the ring approximation we have that

$$\tilde{\chi}_{1/1}(y_1; \epsilon/z) = \tilde{\chi}_{1/1}^{(1)}(y_1; \epsilon/z) + \tilde{\chi}_{1/1}^{(2)}(y_1; \epsilon/z), \tag{30}$$

where $\tilde{\chi}_{1/1}^{(1)}$ is the Boltzmann part

$$\tilde{\chi}_{1/1}^{(1)}(y_1; \epsilon/z) = [\epsilon + L_1^0 - \lambda(1)]^{-1} \delta(y_1 - z) f_{1,\mathrm{eq}}(y_1) \tag{31}$$

and $\tilde{\chi}_{1/1}^{(2)}$ is the contribution coming from the ring events

$$\tilde{\chi}_{1/1}^{(2)}(y_1; \epsilon/z) = [\epsilon + L_1^0 - \lambda(1)]^{-1} \tilde{R}(1; \epsilon) \times [\epsilon + L_1^0 - \lambda(1)]^{-1} \delta(y_1 - z) f_{1,\mathrm{eq}}(y_1). \tag{32}$$

Equations (30)–(32) will be the starting point in the following section.

III. LONG TIME BEHAVIOR OF THE LINEAR BURNETT COEFFICIENTS

We are interested in the long time behavior of correlation functions of the form (3). As all the time dependence is included in the distribution function $\chi_{1/1}(y; \tau/z)$, we are going to study this function. The Boltzmann part $\chi_{1/1}^{(1)}$ is known to decay exponentially over a few mean free times and, in the long time limit that we will consider, can be neglected because we will see that $\chi_{1/1}^{(2)}$ presents a slow decay in time.

Before we proceed further, we make another approximation in Eq. (32) by using point $T^{(0)}(12)$ operators instead of $\overline{T}_-(12)$ operators. The operators $T^{(0)}(12)$ are defined by

$$T^{(0)}(12) = T_0(12)\delta(\mathbf{r}_{12}), \tag{33}$$

$$T_0(12) = \sigma^{d-1} \int_{\mathbf{v}_{12}\cdot\sigma>0} d\hat{\sigma}\, |\mathbf{v}_{12} \cdot \hat{\sigma}|(P_\sigma - 1), \tag{34}$$

i.e., they neglect the difference in position of colliding particles and correspond to a low-density approximation. It has been proved[12] that the use of $T^{(0)}(12)$ operators does not change the behavior of the dominant ring contributions.

It is convenient now to go over to a Fourier representation of the ring operator. By using the Fourier representation of the δ functions we obtain

$$\tilde{\chi}_{1/1}^{(2)}(y_1; \epsilon/z) = \frac{1}{(2\pi)^d} \int d\mathbf{k} \exp[i\mathbf{k} \cdot (\mathbf{r}_1 - \mathbf{r})] \times [\epsilon + i\mathbf{k} \cdot \mathbf{v}_1 - \lambda_0(1)]^{-1} \tilde{R}_0(1; \epsilon) \times [\epsilon + i\mathbf{k} \cdot \mathbf{v}_1 - \lambda_0(1)]^{-1} \delta(\mathbf{p}_1 - \mathbf{p}) f_{1,\mathrm{eq}}(y_1), \tag{35}$$

where

$$\tilde{R}_0(1; \epsilon) = \frac{1}{(2\pi)^d} \int d\mathbf{k}' \int d\mathbf{p}_2\, T_0(12) \times [\epsilon + i\mathbf{k}' \cdot \mathbf{v}_{12} + i\mathbf{k} \cdot \mathbf{v}_1 - \lambda_0(1) - \lambda_0(2)]^{-1} \times T_0(12)(1 + P_{12}) f_{1,\mathrm{eq}}(y_2) \tag{36}$$

and

$$\lambda_0(\alpha) = \int d\mathbf{p}_3\, T_0(\alpha 3)(1 + P_{\alpha 3}) f_{1,\mathrm{eq}}(y_3), \quad \alpha = 1, 2. \tag{37}$$

Here we have used the notation $y_1 \equiv \{\mathbf{r}_1, \mathbf{p}_1\}$, $z \equiv \{\mathbf{r}, \mathbf{p}\}$.

Since we are interested in the long time behavior of $\chi_{1/1}^{(2)}(t)$, we consider $\tilde{\chi}_{1/1}^{(2)}(\epsilon)$ for small values of ϵ. On the other hand, dominant contributions to the k integrals should correspond to small values of $\mathbf{k}$. More precisely, we can neglect contributions coming from values $k > k_0$ when k_0^{-1} is of the order of the mean free path. This is because the contribution from $k > k_0$ part of the integral corresponds to the short time effects taking place on a time scale of the order of a mean free time or less, and we are not interested in them.

To compute the action of the operators appearing in Eq. (35), it is convenient to introduce the eigenvalue problem

$$[i\mathbf{k}\cdot\mathbf{v}_1 - \lambda_0(1)]\Theta^{(\Omega)}(\mathbf{k},\mathbf{v}_1)f_{1,\mathrm{eq}}(y_1) = \Omega(k)\Theta^{(\Omega)}(\mathbf{k},\mathbf{v}_1)f_{1,\mathrm{eq}}(y_1) \tag{38}$$

with the normalization property

$$\int d\mathbf{p}_1\,\Theta^{(\Omega)}(\mathbf{k},\mathbf{v}_1)\Theta^{(\Omega')}(\mathbf{k},\mathbf{v}_1)f_{1,\mathrm{eq}}(y_1) = n\delta_{\Omega,\Omega'}\ . \tag{39}$$

In our study, we will only need[13] the solutions of Eq. (38) corresponding to hydrodynamic modes, i.e., those eigenfunctions which have an eigenvalue going to zero as $k\to 0$. There are two sound modes ($\pm$), one heat mode (T) and $d-1$ shear modes (i; $i=1,2,\ldots,d-1$). Their explicit expressions to the lowest order in k are

$$\Theta_0^{(\pm)}(\hat{\mathbf{k}},\mathbf{v}_1) = \left[\frac{d}{2(d+2)}\right]^{1/2}\left(\frac{\beta m v_1^2}{d} \pm \beta m c_0\,\hat{\mathbf{k}}\cdot\mathbf{v}_1\right),$$

$$\Theta_0^{(T)}(\hat{\mathbf{k}},\mathbf{v}_1) = \left(\frac{2}{d+2}\right)^{1/2}\left(\frac{\beta m v_1^2}{2} - \frac{d+2}{2}\right),$$

$$\Theta_0^{(i)}(\hat{\mathbf{k}},\mathbf{v}_1) = (\beta m)^{1/2}\,\hat{\mathbf{k}}^{(i)}\cdot\mathbf{v}_1 \tag{40}$$

with the hydrodynamic frequencies given by

$$\Omega^{(\pm)}(k) = \pm ikc_0 + \tfrac{1}{2}\Gamma_{s,0}k^2 + O(k^3)\ ,$$

$$\Omega^{(T)}(k) = k^2 D_{T,0} + O(k^4)\ ,$$

$$\Omega^{(i)}(k) = \nu_0 k^2 + O(k^4)\ . \tag{41}$$

Here $\beta = (k_B T)^{-1}$, $(\hat{\mathbf{k}}, \hat{\mathbf{k}}^{(1)}, \ldots, \hat{\mathbf{k}}^{(d-1)})$ are a set of mutually orthogonal unit vectors; $c_0 = (d+2/d\beta m)^{1/2}$ is the adiabatic sound velocity for an ideal gas, $\Gamma_{s,0} = 2(d+1)\nu_0/d + (\gamma_0 - 1)D_{T,0}$ is the sound damping constant, where $\gamma_0 = c_{p,0}/c_{v,0} = (d+2/2)$, $\nu_0 = \eta_0/nm$ and $D_{T,0} = (\lambda_0/nc_{p,0})$; $c_{p,0}$, $c_{v,0}$, η_0, λ_0 being the ideal gas specific heats at constant pressure and at constant volume, the coefficients of viscosity and thermal conductivity from the Boltzmann equation, respectively.

Now, we assume that any function $h(\mathbf{v}_1)$ we can expand in the form

$$h(\mathbf{v}_1) = \sum_{\Omega} c^{(\Omega)}\Theta^{(\Omega)}(\mathbf{k},\mathbf{v}_1)f_{1,\mathrm{eq}}(y_1)\ , \tag{42}$$

where the sum extends over all the eigenfunctions given by Eq. (38). When this spectral decomposition is introduced into Eq. (35), it is seen that in the long time behavior only the hydrodynamic modes have to be considered. The dominant contributions correspond to the limit $k\to 0$, and in the time representation we have factors of

$$\lim_{t\gg}\lim_{k\ll}\exp[-\Omega(k)t]\ .$$

If $\Omega(k)$ is not a hydrodynamic frequency this limit vanishes and its contribution is negligible as compared to those coming from the hydrodynamic ones. Thus, using the espectral decomposition (42) we write Eq. (35) as

$$\begin{aligned}\tilde{\chi}_{1/1}^{(2)}(y_1;\epsilon/z) = {} & \frac{1}{(2\pi)^d}\int d\mathbf{k}\exp[i\mathbf{k}\cdot(\mathbf{r}_1-\mathbf{r})][\epsilon + i\mathbf{k}\cdot\mathbf{v}_1 - \lambda_0(1)]^{-1}\frac{1}{(2\pi)^d}\int d\mathbf{k}'\int d\mathbf{p}_2\,T_0(12)\frac{1}{n^2}\\ & \times\sum_{\Omega}{}'\sum_{\Omega'}{}'\,\Theta_0^{(\Omega)}(\hat{\mathbf{l}},\mathbf{v}_1)\Theta_0^{(\Omega')}(-\hat{\mathbf{k}}',\mathbf{v}_2)f_{1,\mathrm{eq}}(y_1)f_{1,\mathrm{eq}}(y_2)[\epsilon + \Omega(l) + \Omega'(k')]^{-1}\\ & \times\left\{\int d\mathbf{p}_1\int d\mathbf{p}_2\,\Theta_0^{(\Omega)}(\hat{\mathbf{l}},\mathbf{v}_1)\Theta_0^{(\Omega')}(-\hat{\mathbf{k}}',\mathbf{v}_2)T_0(12)(1+P_{12})f_{1,\mathrm{eq}}(y_2)[\epsilon + i\mathbf{k}\cdot\mathbf{v}_1 - \lambda_0(1)]^{-1}\delta(\mathbf{p}_1-\mathbf{p})f_{1,\mathrm{eq}}(y_1)\right\}\ ,\end{aligned} \tag{43}$$

where $\mathbf{l} = \mathbf{k} + \mathbf{k}'$ and the prime on the summation symbol indicates that only hydrodynamic modes should be included in the sum. We have written the lowest order in k of the hydrodynamic modes. Higher order corrections are only needed when the contributions from $\Theta_0^{(\Omega)}$ are zero. This will not be the case in our calculations.

Now, we will introduce expression (43) into Eq. (10) and consider two particular cases denoted by I and II:

$$\text{(I)}\quad F_1(\mathbf{r}_1,\mathbf{p}_1) = F_1(\mathbf{p}_1)\ ,\quad F_1'(\mathbf{r},\mathbf{p}) = F_1'(\mathbf{p})\ , \tag{44}$$

$$\text{(II)}\quad F_1(\mathbf{r}_1,\mathbf{p}_1) = F_1(\mathbf{p}_1)\delta(\mathbf{r}_1)\ ,\quad F_1'(\mathbf{r},\mathbf{p}) = F_1'(\mathbf{p})\mathbf{r}\ . \tag{45}$$

Taking into account Eq. (5) it is clear that the long time behavior of $\rho(\tau)$ is given by

$$\rho(\tau) = \int dy_1\int dz\,F_1(y_1)F_1'(z)\chi_{1/1}^{(2)}(y_1;\tau/z)\ . \tag{46}$$

For case (I) calculations are straightforward. Using the fact that the functions $\Theta_0^{(\Omega)}$ are composed of collision invariants and approximating $[\epsilon - \lambda_0(1)]^{-1}$ by $[-\lambda_0(1)]^{-1}$ we get in the long time limit

$$\rho_{\mathrm{I}}(\tau) = \frac{V}{2n^2}\sum_{\Omega}{}'\sum_{\Omega'}{}'\int\frac{d\mathbf{k}}{(2\pi)^d}\exp\{-\tau[\Omega(k)+\Omega'(k)]\}I_{\Omega,\Omega'}(\hat{\mathbf{k}},F_1)I_{\Omega,\Omega'}(\hat{\mathbf{k}},F_1')\ , \tag{47}$$

where

$$I_{\Omega,\Omega'}[\hat{\mathbf{k}},\phi(\mathbf{p})] = \int d\mathbf{p}\,\phi(\mathbf{p})\Theta_0^{(\Omega)}(\hat{\mathbf{k}},\mathbf{v}_1)\Theta_0^{(\Omega')}(-\hat{\mathbf{k}},\mathbf{v}_1)f_{1,\mathrm{eq}}(y_1)\ . \tag{48}$$

By analyzing the k integral it is seen that contributions of the form $\exp(-atk^2)$ lead to a much more slow damping than those of the form $\exp(-btk)$. Hence only combinations of Ω and Ω' such that $\Omega(k)+\Omega'(k)\sim k^2$ are significant for our calculations. Equation (47) is essentially the result obtained in previous works on the long time tails associated with the Navier–Stokes transport coefficients.[14] In fact, all the correlation functions appearing in the Green–Kubo formula belong to the case I defined by Eq. (44).

In case (II) the factor $\mathbf{r}$ can be written as a derivative with respect to $\mathbf{k}$ of $\exp(-i\mathbf{k}\cdot\mathbf{r})$. After integration by parts, we have the operator $\partial/\partial\mathbf{k}$ acting on the hydrodynamic modes $\Theta_0^{(\Omega)}(1,\mathbf{v}_1)$ and also on the exponential factor $\exp[-\tau\Omega'(l)]$. The dominant contribution comes when acting on the exponential function, bringing down a factor of τ. Using then the same kind of arguments as in case (I) it is found

$$\rho_{II}(\tau)=\int d\mathbf{p}_1\int d\mathbf{p}\,F_1(\mathbf{p}_1)F_1'(\mathbf{p})\frac{1}{(2\pi)^d}\int d\mathbf{k}'\frac{1}{-\lambda_0(1)}\int d\mathbf{p}_2\,T_0(12)\frac{1}{n^2}{\sum_{\Omega}}'{\sum_{\Omega'}}'\Theta_0^{(\Omega)}(\hat{\mathbf{k}}',\mathbf{v}_1)\Theta_0^{(\Omega')}(-\hat{\mathbf{k}}',\mathbf{v}_2)$$
$$\times f_{1,\mathrm{eq}}(y_1)f_{1,\mathrm{eq}}(y_2)\left[-i\frac{\partial\Omega(k')}{\partial k'}\hat{\mathbf{k}}'\right](-\tau)\exp\{-\tau[\Omega(k')+\Omega'(k')]\}\Theta_0^{(\Omega)}(\hat{\mathbf{k}}',\mathbf{v})\Theta_0^{(\Omega')}(-\mathbf{k}',\mathbf{v})f_{1,\mathrm{eq}}(z)\ . \tag{49}$$

From the expressions of the hydrodynamic frequencies it follows that only the derivatives of $\Omega^{(\pm)}$ have contributions of zero order in k. As factors of k lead to more well behaved or less divergent contributions,[15] it follows from this and the comments below Eq. (47) that only the combination of two different sound modes contributes to the long time behavior of Eq. (49). Then, after simple manipulations we get

$$\rho_{II}(\tau)=-\frac{1}{n^2}\tau\int\frac{d\mathbf{k}}{(2\pi)^d}c_0\hat{\mathbf{k}}\exp(-\tau\Gamma_{s,0}k^2)\times I_{+,-}(\hat{\mathbf{k}},F_1)I_{+,-}(\hat{\mathbf{k}},F_1')\ , \tag{50}$$

where the notation (48) has been used again. Now using Eqs. (47) and (50) it is a matter of direct, but tedious, calculations to obtain the behavior of the kinetic part of all the correlation functions appearing in the expressions of the Navier–Stokes transport coefficients and also of the linear Burnett transport coefficients. We list the results including those corresponding to the Navier–Stokes order, that have been already calculated,[5,11] for the sake of completeness.

$$\langle[e^{\tau L}\hat{\mathcal{T}}_{xy}]\hat{\mathcal{T}}_{xy}\rangle=\frac{V\tau^{-d/2}}{2^d\pi^{d/2}\beta^2d(d+2)}\left[\Gamma_{s,0}^{-d/2}+(2\nu_0)^{-d/2}(d^2-2)\right]\ , \tag{51}$$

$$\langle[e^{\tau L}\hat{\mathcal{J}}_i^E]\hat{\mathcal{J}}_j^E\rangle=\frac{V(d+2)\tau^{-d/2}}{2^d\pi^{d/2}\beta^3md}\times\left[\frac{1}{d}\Gamma_{s,0}^{-d/2}+\frac{d-1}{2}(\nu_0+D_{T,0})^{-d/2}\right]\delta_{ij}\ , \tag{52}$$

$$\int d\mathbf{r}'\,\langle[e^{\tau L}\hat{\tau}_{ij}(0)]\hat{J}_k^E(\mathbf{r}')\rangle r_l'=-\frac{\Gamma_{s,0}^{-d/2}\tau^{1-d/2}}{2^d\pi^{d/2}\beta^3md^2}\left(-\frac{2}{d}\delta_{ij}\delta_{kl}+\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}\right)\ . \tag{53}$$

As it is well known the correlation function associated with the bulk viscosity ζ identically vanishes in the kinetic approximation. Also, only one of the two transport coefficients associated with the left-hand side of Eq. (53) remains in the approximation we are considering. From the definitions of K_1 and K_2 in Eq. (A7) and the expression (53) it follows that $K_2+(2/d)K_1=0$.

So, for the transport coefficients as defined in the Appendix A and in Eq. (1) we have

$$\eta\sim\frac{1}{2^d\pi^{d/2}\beta d(d+2)}\times\left[\Gamma_{s,0}^{-d/2}+(2\nu_0)^{-d/2}(d^2-2)\right]\int_0^\infty d\tau\,\tau^{-d/2}\ , \tag{54a}$$

$$\lambda\sim\frac{k_B(d+2)}{2^d\pi^{d/2}\beta md}\left[\frac{1}{d}\Gamma_{s,0}^{-d/2}+\frac{d-1}{2}(\nu_0+D_{T,0})^{-d/2}\right]\int_0^\infty d\tau\,\tau^{-d/2}\ , \tag{54b}$$

$$\eta_1\sim\frac{1}{2^d\pi^{d/2}\beta d(d+2)}\times\left[\Gamma_{s,0}^{-d/2}+(2\nu_0)^{-d/2}(d^2-2)\right]\int_0^\infty d\tau\,\tau^{1-d/2}\ , \tag{54c}$$

$$\eta_2\sim-\frac{2}{d}\eta_1\ , \tag{54d}$$

$$\eta_3\sim\frac{k_B\Gamma_{s,0}^{-d/2}\pi^{-d/2}}{2^{d-1}d^2\beta m}\int_0^\infty d\tau\,\tau^{1-d/2}\ , \tag{54e}$$

$$\eta_4\sim-\frac{1}{d}\eta_3\ , \tag{54f}$$

$$\lambda_1\sim\frac{k_B(d+2)}{2^d\pi^{d/2}\beta md}\left[\frac{1}{d}\Gamma_{s,0}^{-d/2}+\frac{d-1}{2}(\nu_0+D_{T,0})^{-d/2}\right]\int_0^\infty d\tau\,\tau^{1-d/2}\ , \tag{54g}$$

$$\lambda_2=\frac{T}{2}(\eta_3+2\eta_4)\ , \tag{54h}$$

$$\lambda_3=\tfrac{1}{2}T\eta_3\ . \tag{54i}$$

Upon writing these expressions we have formally assumed that the time behavior described by Eqs. (51)–(53) extends over arbitrarily long times. The expressions (54a) and (54b) have been derived many times by using different kinds of theories and, in particular, the one we have followed in this paper.[5]

The long time behavior of the linear Burnett transport coefficients has been studied in Refs. 7 and 16. Ernst and Dorfman[7] use mode–mode coupling theory to evaluate the dominant contributions. Although the low density limit of their results and ours are consistent, they cannot be said to be equivalent because Ernst and Dorfman only consider some combinations of the transport coefficients (54c)–(54i). An outline of the comparison of both results is given in Appendix B. Sharma[16] starts from the nonlinear kinetic equation derived by Dorfman and Cohen[10] and uses kinetic theory to evaluate the dominant long time form of the hydrodynamic equations. His results are equivalent to Eqs. (54).

We have shown, therefore, that all linear Burnett coefficients seem to diverge in two and three dimensions. More explicitly, they are expressed as the limit for $t\to\infty$ of time integrals of quantities that for long times behave as constants in two dimensions and as $t^{-1/2}$ in three dimensions.

IV. NONLINEAR BURNETT TRANSPORT COEFFICIENTS

In this section we will discuss the long time behavior of the nonlinear Burnett coefficients, i.e., of the coefficients of terms of the form $(\nabla B)(\nabla B)$ in Eqs. (A1) and (A2). Here B denotes any of the local thermodynamic variables describing the system. A look to Eq. (A7) shows that the nonlinear Burnett transport coefficients are related to several types of correlation functions. In the kinetic approximation, if we want to cover all the possible cases we have to consider, in addition to those studied in the previous section, the general expression

$$\mu(\tau)=\langle F(\tau)F'(0)F''(0)\rangle\ , \tag{55}$$

where we have used the same notation as in Eq. (3) and F'' has the same form as F and F'. We notice by passing that we only need one-time correlation functions, although they involve three phase functions, but not two-time correlation functions.

The method developed in Sec. III can be easily generalized to expressions like Eq. (55). We define a distribution function

$$\begin{aligned}K_2(x_1,\ldots,x_N;\tau/z_1,z_2)&=\exp[-\tau L(x)]\Big[\sum_{\alpha_1}\sum_{\substack{\alpha_2\\ \alpha_1\neq\alpha_2}}\delta(z_1-x_{\alpha_1})\\&\qquad\times\delta(z_2-x_{\alpha_2})f_{\rm eq}(x_1,\ldots,x_N)\Big]\\&=\sum_{\alpha_1}\sum_{\substack{\alpha_2\\ \alpha_1\neq\alpha_2}}\delta[z_1-x_{\alpha_1}(-\tau)]\delta[z_2-x_{\alpha_2}(-\tau)]f_{\rm eq}(x_1,\ldots,x_N)\end{aligned} \tag{56}$$

and a set of reduced distributions

$$\begin{aligned}f_{s/2}(y_1,\ldots,y_s;\tau/z_1,z_2)&=\sum_{\alpha_1}\cdots\sum_{\substack{\alpha_s\\ \alpha_1\neq\cdots\neq\alpha_s}}\int dx_1\ldots dx_N\,\delta(y_1-x_{\alpha_1})\ldots\delta(y_s-x_{\alpha_s})\\&\quad\times K_2(x_1,\ldots,x_N;\tau/z_1,z_2)\ .\end{aligned} \tag{57}$$

It is then seen that $\mu(\tau)$ can be expressed as

$$\begin{aligned}\mu(\tau)&=\int dy_1\int dz_1\int dz_2\,F_1(y_1)F_1'(z_1)F_1''(z_2)f_{1/2}(y_1;\tau/z_1,z_2)\\&\quad+\int dy_1\int dz_1\,F_1(y_1)F_1'(z_1)F_1''(z_1)f_{1/1}(y_1;\tau/z_1)\ ,\end{aligned} \tag{58}$$

where $f_{1/1}$ was defined in Eq. (9). We introduce cluster functions by the expansion

$$\begin{aligned}f_{1/2}(y_1;\tau/z_1,z_2)&=f_{1,\rm eq}(y_1)f_{1,\rm eq}(z_1)f_{1,\rm eq}(z_2)\\&\quad+f_{1,\rm eq}(y_1)g_{2,\rm eq}(z_1,z_2)+f_{1,\rm eq}(z_1)\chi_{1/1}(y_1;\tau/z_2)\\&\quad+f_{1,\rm eq}(z_2)\chi_{1/1}(y_1;\tau/z_1)+\chi_{1/2}(y_1;\tau/z_1,z_2)\end{aligned} \tag{59}$$

and similarly for $f_{2/2}$, etc. These relations define $\chi_{s/2}$ once the $\chi_{s/1}$ have been defined in Eq. (16). Taking into account the property (5) we can rewrite Eq. (58) as

$$\begin{aligned}\mu(\tau)&=\int dy_1\int dz_1\int dz_2\,F_1(y_1)F_1'(z_1)F_1''(z_2)\chi_{1/2}(y_1;\tau/z_1,z_2)\\&\quad+\int dy_1\int dz_1\,F_1(y_1)F_1'(z_1)F_1''(z_1)\chi_{1/1}(y_1;\tau/z_1)\ .\end{aligned} \tag{60}$$

From the above definitions it can be derived a hierarchy of equations for the correlation functions $\chi_{s/2}(\tau)$. For the case of hard spheres or disks the two first equations of the hierarchy are

$$[\partial_\tau+L_1^0-\lambda(1)]\chi_{1/2}(y_1;\tau/z_1,z_2)=\lambda(12)\chi_{1/1}(y_1;\tau/z_1)\chi_{1/1}(y_2;\tau/z_2)+\int dy_2\,\overline{T}_-(12)\chi_{2/2}(y_1,y_2;\tau/z_1,z_2)\ , \tag{61a}$$

$$\begin{aligned}&[\partial_\tau+L_1^0+L_2^0-\overline{T}_-(12)-\Lambda(12)]\chi_{2/2}(y_1,y_2;\tau/z_1,z_2)\\&=\overline{T}_-(12)(1+P_{12})[\chi_{1/1}(y_1;\tau/z_1)\chi_{1/1}(y_2;\tau/z_2)+\chi_{1/2}(y_1;\tau/z_1,z_2)f_{1,\rm eq}(y_2)]+(1+P_{12})\int dy_3\,\overline{T}_-(13)\{(1+P_{13})[g_{2,\rm eq}(y_1,y_2)\\&\quad\times\chi_{1/2}(y_3;\tau/z_1,z_2)+\chi_{1/1}(y_1;\tau/z_1)\chi_{2/1}(y_2,y_3;\tau/z_2)+\chi_{1/1}(y_1;\tau/z_2)\chi_{2/1}(y_2,y_3;\tau/z_1)]+\chi_{3/2}(y_1,y_2,y_2;\tau/z_1,z_2)\}\ ,\end{aligned} \tag{61b}$$

where we have introduced the nonlinear operator

$$\lambda(12)=\int dy_2\,\overline{T}_-(12)(1+P_{12})\ , \tag{62}$$

the other symbols being the same as in Sec. II. The initial conditions for Eqs. (61) are obtained by comparison of the definition (57) and the expansion (59) for $\tau=0$. The value $\chi_{1/1}(\tau=0)$ is given in Eq. (22). In this way one gets

$$\begin{aligned}\chi_{1/2}(y_1;0/z_1,z_2)&=g_{2,\rm eq}(z_1,z_2)[\delta(y_1-z_1)+\delta(y_1-z_2)]\\&\quad+g_{3,\rm eq}(y_1,z_1,z_2)\ .\end{aligned} \tag{63}$$

The explicit expression of $\chi_{2/2}(\tau=0)$ will not be given here, but it involves at least three-particle equilibrium correlation functions.

Three-particle correlation functions very similar to $\chi_{s/2}$ have been considered before[18] for a low-density gas, and the results have been used to compute correlation functions appearing in the expressions of nonlinear Burnett coefficients.[8,19] A comparison of the low-density limit of Eqs. (61) with the results obtained by Dufty is given in the Appendix C. Here we will study the ring approximation. Keeping exactly the same kind of dynamical events as in Sec. II, Eqs. (61) yield in the Laplace representation

$$\tilde\chi_{1/2}(y_1;\epsilon/z_1,z_2)=\sum_{i=1}^{5}\tilde\chi_{1/2}^{(i)}(y_1;\epsilon/z_1,z_2)\ , \tag{64}$$

with

$$\tilde\chi_{1/2}^{(1)}(y_1;\epsilon/z_1,z_2)=[\epsilon+L_1^0-\lambda(1)]^{-1}\chi_{1/2}(y_1;0/z_1,z_2)\ , \tag{65}$$

$$\begin{aligned}&\tilde\chi_{1/2}^{(2)}(y_1;\epsilon/z_1,z_2)\\&=[\epsilon+L_1^0-\lambda(1)]^{-1}\lambda(12)\tilde\Xi(y_1,y_2;\epsilon/z_1,z_2)\ ,\end{aligned} \tag{66}$$

$$\tilde{\chi}_{1/2}^{(3)}(y_1;\epsilon/z_1,z_2)=[\epsilon+L_1^0-\lambda(1)]^{-1}\times\tilde{R}(1;\epsilon)[\epsilon+L_1^0-\lambda(1)]^{-1}\chi_{1/2}(y_1;0/z_1,z_2)\ , \tag{67}$$

$$\tilde{\chi}_{1/2}^{(4)}(y_1;\epsilon/z_1,z_2)=[\epsilon+L_1^0-\lambda(1)]^{-1}\times\tilde{R}(1;\epsilon)[\epsilon+L_1^0-\lambda(1)]^{-1}\lambda(12)\tilde{\Xi}(y_1,y_2;\epsilon/z_1,z_2)\ , \tag{68}$$

$$\tilde{\chi}_{1/2}^{(5)}(y_1;\epsilon/z_1,z_2)=[\epsilon+L_1^0-\lambda(1)]^{-1}\tilde{R}'(1;\epsilon)\tilde{\Xi}(y_1,y_2;\epsilon/z_1,z_2)\ . \tag{69}$$

In these expressions $\tilde{R}(1;\epsilon)$ is the ring operator defined by Eq. (29) and

$$R'(1;\epsilon)=\int dy_2\,\overline{T}_-(12)\times[\epsilon+L_1^0+L_2^0-\,\hat{}\,(12)]^{-1}\,\overline{T}_-(12)(1+P_{12})\ . \tag{70}$$

We have also introduced the auxiliary quantity $\tilde{\Xi}(y_1,y_2;\epsilon/z_1,z_2)$ that is the Laplace transform of

$$\Xi(y_1,y_2;\tau/z_1,z_2)=\chi_{1/1}(y_1;\tau/z_1)\chi_{1/1}(y_2;\tau/z_2)\ . \tag{71}$$

We will need the expression of Ξ to the lowest order in the density and that, according to Eq. (31), it is

$$\tilde{\Xi}(y_1,y_2;\epsilon/z_1,z_2)=[\epsilon+L_1^0+L_2^0-\,\hat{}\,(12)]^{-1}\times\delta(y_1-z_1)\delta(y_2-z_2)f_{1,\mathrm{eq}}(y_1)f_{1,\mathrm{eq}}(y_2)\ . \tag{72}$$

Let us analyze each of the five terms contributing to $\chi_{1/2}$. The first term $\chi_{1/2}^{(1)}$ can be neglected in the long time limit as it yields an exponential decay. The second term $\chi_{1/2}^{(2)}$ contains three kinds of contributions depending on the approximation considered for Ξ. We distinguish the three following cases. (1) $\chi_{1/1}(y_1;\tau/z_1)$ and $\chi_{1/1}(y_2;\tau/z_2)$ are considered to Boltzmann's order. It is clear that we have for $\Xi(\tau)$, and also for $\chi_{1/2}^{(2)}(\tau)$ an exponential decay. (2) $\chi_{1/1}(y_1;\tau/z_1)$ is considered in the ring approximation and $\chi_{1/1}(y_2;\tau/z_2)$ in the Boltzmann approximation. For long enough times we also have an exponential decay. Of course, the same applies to the opposite case. (3) Both $\chi_{1/1}(y_1;\tau/z_1)$ and $\chi_{1/1}(y_2;\tau/z_2)$ are considered in the ring approximation. Using the results in Sec. III it is then seen that we obtain contributions to $\mu(\tau)$ that are less divergent than those arising from the second term on the right-hand side of Eq. (58). To be more precise, if the latter gives contributions depending on time as $\tau^{1-d/2}$, we get in this approximation contributions that are proportional to τ^{1-d}, thus having a better long time behavior.

The only difference between the expression (67) of $\tilde{\chi}_{1/2}^{(3)}$ and the expression (32) of $\tilde{\chi}_{1/1}^{(2)}$ lies in the initial value. In Eq. (67) the initial value is given by Eq. (63) while in Eq. (32) it is $\delta(y_1-z)f_{1,\mathrm{eq}}(y_1)$. Taking into account that we can write

$$\int dy_1\int dz_1\,F_1(y_1)F_1'(z_1)F_1''(z_1)\chi_{1/1}(y_1;\tau/z_1)=\int dy_1\int dz_1\int dz_2\,F_1(y_1)F_1'(z_1)F_1''(z_2)\times\chi_{1/1}(y_1;\tau/z_1)\delta(z_1-z_2)\ , \tag{73}$$

it is clear that contributions coming from this term in the ring approximation are of lower order in density than those coming from Eq. (67), which in this way must also be neglected by consistency.

The analysis of $\chi_{1/2}^{(4)}$ and $\chi_{1/2}^{(5)}$ is much more complicated. As the arguments go in the same line as in Sec. III we only quote the final result here and we will sketch the proof in Appendix D. Contributions to $\mu(\tau)$ coming from $\chi_{1/2}^{(4)}$ and $\chi_{1/2}^{(5)}$ exactly cancel each other, at least in the cases that are relevant for the calculation of nonlinear Burnett transport coefficients. Of course, this remarkable property is only true as long as to the leading contributions to the long time behavior is concerned.

The final conclusion of our discussion is that only contributions associated to $\chi_{1/1}(y_1;\tau/z_1)$ must be kept in our approximation. The long time behavior of Eq. (60) is given by

$$\mu(\tau)=\int dy_1\int dz_1\,F_1(y_1)F_1'(z_1)F_1''(z_1)\chi_{1/1}^{(2)}(y_1;\tau/z_1)\ . \tag{74}$$

Of course, all the analysis carried out in the previous section is valid here. In particular, we can use expressions (47) or (50), depending on the form of the product $F_1'(z_1)F_1''(z_1)$. As a direct consequence of this, all the expressions of the nonlinear Burnett coefficients are going to diverge. More precisely, they diverge in exactly the same way as the linear ones, i.e., they are time integrals of functions behaving for long times as $\tau^{1-d/2}$, where d is the dimension of the system.

We will not intend here to give the explicit expression of the long time behavior of all the nonlinear Burnett coefficients. We will limit ourselves, because of their special interest, to the so-called viscometric functions characterizing normal stresses in a fluid in steady shear flow, with uniform temperature and pressure. We assume that the only nonvanishing component of the flow field is along the x axis

$$u_x(\mathbf{r})=ar_y\ , \tag{75}$$

where a is a constant, characterizing the size of the velocity gradient. The viscometric functions $\Psi_1(a)$ and $\Psi_2(a)$ are defined in terms of the dissipative part of the stress tensor $\langle\tau_{ij}\rangle^{(D)}$ as[17]

$$\langle\tau_{xx}\rangle^{(D)}-\langle\tau_{yy}\rangle^{(D)}=-a^2\Psi_1(a)\ ,$$
$$\langle\tau_{yy}\rangle^{(D)}-\langle\tau_{zz}\rangle^{(D)}=-a^2\Psi_2(a)\ . \tag{76}$$

The Eq. (A1) when particularized for the physical situation we are considering allows us to identify $\Psi_1(0)$ and $\Psi_2(0)$:

$$\Psi_1(0)=\eta_{13}\ ,$$
$$\Psi_2(0)=-\frac{\eta_{12}}{4}-\frac{\eta_{13}}{2}\ , \tag{77}$$

or in terms of correlation functions

$$\Psi_2(0)=\frac{2K_{11}}{k_B^2T^2}-\frac{2K_{20}}{k_BT}\ , \tag{78}$$

$$\Psi_1(0)=-\Psi_2(0)+\frac{2K_{12}}{k_B^2T^2}\ , \tag{79}$$

where

$$K_{11}=\int_0^\infty d\tau\int d\mathbf{r}'\,\langle[e^{\tau L}\hat{\tau}_{zx}(0)]\hat{T}_{xy}G_y(\mathbf{r}')\rangle r_z'\ , \tag{80}$$

$$K_{12}=\int_0^\infty d\tau\int d\mathbf{r}'\,\langle[e^{\tau L}\hat{\tau}_{zx}(0)]\hat{T}_{xy}G_z(\mathbf{r}')\rangle r_y'\ , \tag{81}$$

$$K_{20}=\int_0^\infty d\tau \int d\mathbf{r}' \langle [e^{\tau L}\hat{\tau}_{zx}(0)]G_x(\mathbf{r}')\rangle r'_z \; . \tag{82}$$

Using now Eq. (50) we can compute the long time behavior of the kinetic part of the correlation functions appearing in these equations. The result is

$$\int d\mathbf{r}' \langle [e^{\tau L}\hat{\tau}_{ij}(0)]\hat{T}_{kl}\, G_m(\mathbf{r}')\rangle r'_n = -\frac{1}{\beta^3}\frac{d+4}{d^3}(4\pi\Gamma_{s,0})^{-d/2}\tau^{1-d/2}\left\{\frac{4}{d(d+2)}\delta_{ij}\delta_{kl}\delta_{mn}-\frac{2}{d+2}\delta_{ij}(\delta_{km}\delta_{ln}+\delta_{kn}\delta_{lm})\right.$$
$$\left. -\frac{2}{d+2}\delta_{kl}(\delta_{im}\delta_{nj}+\delta_{in}\delta_{jm})+\frac{d}{d+2}[\delta_{km}(\delta_{il}\delta_{jn}+\delta_{in}\delta_{jl})+\delta_{lm}(\delta_{ik}\delta_{jn}+\delta_{jk}\delta_{in})]\right\} \tag{83}$$

and

$$\int d\mathbf{r}' \langle [e^{\tau L}\hat{\tau}_{ij}(0)]G_k(\mathbf{r}')\rangle r'_l$$
$$=\frac{\tau^{1-d/2}}{2^d\pi^{d/2}\beta^2 d^2}(\Gamma_{s,0})^{-d/2}\left[\frac{2}{d}\delta_{ij}\delta_{kl}-(\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk})\right]. \tag{84}$$

Substitution in Eqs. (78) and (79) yields

$$\Psi_2(0)=-\frac{4}{(d+2)d^2\beta}(4\pi\Gamma_{s,0})^{-d/2}\int_0^\infty d\tau\, \tau^{1-d/2}\;, \tag{85}$$

$$\Psi_1(0)=-\Psi_2(0)\; . \tag{86}$$

The correlation function giving K_{12} vanishes in our approximation. So, the expression of the viscometric functions are also divergent, in the sense we have already discussed, and no casual cancellation takes place.

The viscometric functions as given by Eqs. (78) and (79) have been evaluated at low density for Maxwell molecules by Dufty and Marchetti.[8] Of course, they obtain an exponential decay for the correlation function. Nevertheless, it must be pointed out that the symmetry of their results is different from the one presented here, in the sense that they get $\Psi_2(0)=0$ but $\Psi_1(0)\neq 0$.

V. DISCUSSION

In this paper we have considered the long time behavior of the correlation functions that appear in the Green–Kubo-like formula of the Burnett transport coefficients. We have used renormalized kinetic theory, taking into account only the Boltzmann term and the contributions from the three body and all higher body ring events. The correlation functions present hydrodynamic tails in such a way that all the Burnett coefficients, linear as well as nonlinear, diverge as $t^{2-d/2}$ in time, where d is the dimension of the system. The coefficients of the $t^{1/2}$ term in the linear Burnett transport coefficients for $d=3$ are in agreement with the results obtained by Ernst and Dorfman using mode–mode coupling theory.[7]

We have confined our calculations to the kinetic parts of the time correlation functions and to low densities. So, we have ignored, for instance, the Enskog type excluded volume corrections. Nevertheless, the results may be generalized following the method developed by Cohen and Dorfman.[20]

Although we only know the explicit expressions of the transport coefficients up to Burnett order, their general structure to an arbitrary order in the gradients can be easily guessed. From the formal derivations of transport equations[2,3] it follows that all the transport coefficients can be expressed as space and time moments of one time correlation functions. These correlations functions involve a number of dynamical variables that depends on the nonlinearity of the associated term in the transport equations. Nevertheless, this fact does not affect the long time behavior of the transport coefficients. What is relevant is the number of time and position factors that are present in their expression, and this number is determined by the number of gradient operators to its right in the transport equation. Each position factor implies a time factor in the long time limit. As a consequence, the expressions of all the transport coefficients to a given order in gradients will diverge in the same way. By passing, we notice that the presence of position factors gives a special role to the sound hydrodynamic modes as long as the long time behavior is concerned.

The main implication of the apparent divergences of the Burnett and higher order coefficients in three dimensions and of the Navier–Stokes and higher order coefficients in two dimensions is that the structure of hydrodynamic equations must be reexaminated. In particular, expansions of the pressure tensor and the heat current in powers of the gradients of the hydrodynamic variables must be avoided. Some progress in this direction has been made by using mode–mode coupling theory and also kinetic theory.[6] Nevertheless, a great deal of work remains to be done in this field.

We want to point out that in spite of the problem of the divergencies, an expansion in gradients of the transport equations may still be useful. First, it may allow the calculation of the "regular" or nonsingular part of these coefficients and this may be interesting since the regular part is probably dominant for small, but not asymptotically small, gradients. Second, the formal expansions can be used to establish the range of validity of some phenomenological relations. An example of this will be published in the near future.

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APPENDIX A: HYDRODYNAMIC EQUATIONS TO BURNETT ORDER FOR ONE-COMPONENT FLUIDS

Here we present the result one gets when the formal equation (74) in Ref. 3 is applied to the case of simple one-component fluids. The dissipative part of the momentum flux is given by

$$\begin{aligned}\langle\tau_{ij}(\mathbf{r})\rangle_t^{(D)} = & -\eta\left(\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}-\frac{2}{d}\delta_{ij}\delta_{kl}\right)\frac{\partial}{\partial r_l}u_k-\zeta\delta_{ij}\nabla\cdot\mathbf{u}-\frac{2\eta_1}{mn}\left(\frac{\partial^2 p}{\partial r_i\partial r_j}-\kappa_T\frac{\partial p}{\partial r_i}\frac{\partial p}{\partial r_j}\right)+\left(\eta_7-\frac{\alpha\eta_1}{mn}\right)\left(\frac{\partial p}{\partial r_i}\frac{\partial T}{\partial r_j}+\frac{\partial p}{\partial r_j}\frac{\partial T}{\partial r_i}\right)\\ & +\left[\eta_{11}-2n\left(\frac{\partial\eta_1}{\partial n}\right)_s\right]\nabla\cdot\mathbf{u}D_{ij}+\delta_{ij}\left\{\eta_4\nabla^2T+\eta_6(\nabla T)^2-\frac{\eta_2}{mn}[\nabla^2p-\kappa_T(\nabla p)^2]+\left[\eta_8-\frac{\alpha\eta_2}{mn}\right]\nabla p\cdot\nabla T+(\eta_{10}-\eta_2)D_{kl}D_{kl}\right.\\ & \left.+\eta_2w_{kl}w_{kl}+\left[\eta_9-n\left(\frac{\partial\eta_2}{\partial n}\right)_s\right](\nabla\cdot\mathbf{u})^2\right\}+\eta_3\frac{\partial^2T}{\partial r_i\partial r_j}+\eta_5\frac{\partial T}{\partial r_i}\frac{\partial T}{\partial r_j}+(\eta_{12}-2\eta_1)D_{ik}D_{jk}+\eta_{13}(D_{ki}w_{kj}+D_{kj}w_{ki})+2\eta_1w_{ik}w_{jk}.\end{aligned}\tag{A1}$$

The dissipative part of the heat flux is given by

$$\begin{aligned}\langle J_i^E(\mathbf{r})\rangle_t^{(D)} = & u_j\langle\tau_{ij}(\mathbf{r})\rangle_t^{(D)}-\lambda\frac{\partial}{\partial r_i}T+\left[\lambda_2-\lambda_1T\left(\frac{\partial p}{\partial e}\right)_n\right]\frac{\partial}{\partial r_i}\nabla\cdot\mathbf{u}+\lambda_3\nabla^2u_i+\left\{\lambda_4-\lambda_1\left(\frac{\partial p}{\partial e}\right)_n-\lambda_1T\left[\frac{\partial}{\partial T}\left(\frac{\partial p}{\partial e}\right)_n\right]_p\right.\\ & \left.-n\left(\frac{\partial\lambda_1}{\partial n}\right)_s\right\}\frac{\partial T}{\partial r_i}\nabla\cdot\mathbf{u}+(\lambda_5-\lambda_1)D_{ij}\frac{\partial T}{\partial r_j}+(\lambda_6+\lambda_1)w_{ij}\frac{\partial T}{\partial r_j}+\lambda_7D_{ij}\frac{\partial p}{\partial r_j}+\left\{\lambda_8-\lambda_1T\left[\frac{\partial}{\partial p}\left(\frac{\partial p}{\partial e}\right)_n\right]_T\right\}\frac{\partial p}{\partial r_i}\nabla\cdot\mathbf{u}.\end{aligned}\tag{A2}$$

In these expressions d is the dimension of the system, r_i is the i component of the position vector $\mathbf{r}$, T is the temperature, p is the pressure, n is the number of particles density, m is the mass of the particles, $\mathbf{u}$ the macroscopic velocity, $\alpha=(-1/n)(\partial n/\partial T)_p$ is the expansion coefficient, $\kappa_T=(1/n)(\partial n/\partial p)_T$ is the isothermal compressibility, e is the internal energy density, s is the entropy density, and w_{ij} and D_{ij} are the vorticity and strain rate defined by

$$w_{ij}=\frac{1}{2}\left(\frac{\partial u_i}{\partial r_j}-\frac{\partial u_j}{\partial r_i}\right),\tag{A3}$$

$$D_{ij}=\frac{1}{2}\left(\frac{\partial u_i}{\partial r_j}+\frac{\partial u_j}{\partial r_i}\right).\tag{A4}$$

All these quantities are evaluated at position $\mathbf{r}$ and time t. The coefficients η's and λ's are defined in terms of correlations functions. For the sake of simplicity we write them first as a function of some auxiliary quantities K_i and then give the expressions for these quantities

$$\begin{aligned}\eta&=\frac{1}{k_BT}K^{\mathrm{I}},\\ \zeta&=\frac{1}{k_BT}\left(K^{\mathrm{II}}+\frac{2}{d}K^{\mathrm{I}}\right),\\ \eta_1&=\frac{1}{k_BT}K^{\mathrm{IV}},\\ \eta_2&=\frac{1}{k_BT}K^{\mathrm{V}},\\ \eta_3&=-\frac{2}{k_BT^2}K_1,\\ \eta_4&=-\frac{1}{k_BT^2}K_2,\\ \eta_5&=\frac{4K_1}{k_BT^3}+2K_3\frac{h}{k_B^2T^4n}-\frac{2K_5}{k_B^2T^4}+\frac{2}{k_BT^2}\left(\frac{\partial}{\partial T}\frac{h}{mn}\right)_pK_{20},\\ \eta_6&=\frac{2K_2}{k_BT^3}+K_4\frac{h}{k_B^2T^4n}-\frac{K_6}{k_B^2T^4}+\frac{1}{k_BT^2}\left(\frac{\partial}{\partial T}\frac{h}{mn}\right)_pK_{21},\\ \eta_7&=-\frac{1}{k_B^2T^3n}K_3+\frac{1}{k_BT^2}\left(\frac{\partial}{\partial p}\frac{h}{mn}\right)_TK_{20},\\ \eta_8&=-\frac{1}{k_B^2T^3n}K_4+\frac{1}{k_BT^2}\left(\frac{\partial}{\partial p}\frac{h}{mn}\right)_TK_{21},\\ \eta_9&=-\frac{1}{k_B^2T^2}K_7-\frac{1}{k_BT}\left(\frac{\partial p}{\partial e}\right)_nK_{21},\\ \eta_{10}&=-\frac{2}{k_BT^2}K_8+\frac{2}{k_BT}K_{21},\\ \eta_{11}&=-\frac{2}{k_B^2T^2}K_9-\frac{2}{k_B^2T^2}K_{10}-\frac{2}{k_BT}\left(\frac{\partial p}{\partial e}\right)_nK_{20},\\ \eta_{12}&=-\frac{4}{k_B^2T^2}K_{11}-\frac{4}{k_B^2T^2}K_{12}+\frac{4}{k_BT}K_{20},\\ \eta_{13}&=-\frac{2}{k_B^2T^2}K_{11}+\frac{2}{k_B^2T^2}K_{12}+\frac{2}{k_BT}K_{20},\end{aligned}\tag{A5}$$

and

$$\begin{aligned}\lambda&=\frac{K^{\mathrm{III}}}{k_BT^2},\\ \lambda_1&=\frac{1}{k_BT^2}K^{\mathrm{VI}},\\ \lambda_2&=-\frac{1}{k_BT}(K_1+K_2)=\frac{T}{2}(\eta_3+2\eta_4),\\ \lambda_3&=-\frac{K_1}{k_BT}=\tfrac{1}{2}T\eta_3,\\ \lambda_4&=\frac{1}{k_BT^2}\left\{K_1+K_2-\frac{K_{19}}{k_BT}-\frac{K_{13}}{k_BT}+\frac{h}{nk_BT}K_{17}\right.\\ &\qquad\left.+T\left[\frac{\partial}{\partial T}\left(\frac{\partial p}{\partial n}\right)_e\right]_pK_{22}+T\left[\frac{\partial}{\partial T}\left(\frac{\partial p}{\partial e}\right)_n\right]_pK_{23}\right\},\\ \lambda_5&=\frac{1}{k_BT^2}\left\{3K_1+K_2+\frac{2h}{k_BTn}K_{16}-\frac{2}{k_BT}K_{18}-\frac{1}{k_BT}K_{14}\right.\\ &\qquad\left.-\frac{1}{k_BT}K_{15}+\left[1+\left(\frac{\partial p}{\partial e}\right)_n\right]K_{23}+\left[\left(\frac{\partial p}{\partial n}\right)_e-\frac{h}{n}\right]K_{22}\right\},\\ \lambda_6&=\frac{1}{k_BT^2}\left\{K_1-K_2-\frac{K_{14}}{k_BT}+\frac{K_{15}}{k_BT}\right.\\ &\qquad\left.-\left[1+\left(\frac{\partial p}{\partial e}\right)_n\right]K_{23}-\left[\left(\frac{\partial p}{\partial n}\right)_e-\frac{h}{n}\right]K_{22}\right\},\end{aligned}$$

$$\lambda_7 = -\frac{2}{k_B^2 T^2 n} K_{16} - \frac{2}{mn}\eta_1 ,$$

$$\lambda_8 = -\frac{1}{k_B^2 T^2 n} K_{17} - \frac{1}{mn}\eta_2 + \frac{1}{k_B T}\left[\frac{\partial}{\partial p}\left(\frac{\partial p}{\partial n}\right)_e\right]_T K_{22} + \frac{1}{k_B T}\left[\frac{\partial}{\partial p}\left(\frac{\partial p}{\partial e}\right)_n\right]_T K_{23} \tag{A6}$$

where k_B is the Boltzmann constant and $h = e + p$ is the enthalpy per unit volume. The K's are related to correlation functions through the relations

$$\int_0^\infty ds\, \langle [e^{sL}\hat{\tau}_{ij}(\mathbf{r})]\hat{\mathcal{T}}_{kl}\rangle = K^{\rm I}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + K^{\rm II}\delta_{ij}\delta_{kl} ,$$

$$\int_0^\infty ds\, \langle [e^{sL}\hat{J}_i^E(\mathbf{r})]\hat{\mathcal{J}}_j^E\rangle = K^{\rm III}\delta_{ij} ,$$

$$\int_0^\infty ds\, s\, \langle [e^{sL}\hat{\tau}_{ij}(\mathbf{r})]\hat{\mathcal{T}}_{kl}\rangle = K^{\rm IV}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + K^{\rm V}\delta_{ij}\delta_{kl} ,$$

$$\int_0^\infty ds\, s\, \langle [e^{sL}\hat{J}_i^E(\mathbf{r})]\hat{\mathcal{J}}_j^E\rangle = K^{\rm VI}\delta_{ij} ,$$

$$\int_0^\infty ds \int d\mathbf{r}'\, \langle [e^{sL}\hat{\tau}_{ij}(0)]\hat{J}_k^E(\mathbf{r}')\rangle r'_l = K_1(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + K_2\delta_{ij}\delta_{kl} ,$$

$$\int_0^\infty ds \int d\mathbf{r}'\, \langle [e^{sL}\hat{\tau}_{ij}(0)]\hat{\mathcal{J}}_k^E N'(\mathbf{r}')\rangle r'_l = K_3(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + K_4\delta_{ij}\delta_{kl} ,$$

$$\int_0^\infty ds \int d\mathbf{r}'\, \langle [e^{sL}\hat{\tau}_{ij}(0)]\hat{\mathcal{J}}_k^E E'(\mathbf{r}')\rangle r'_l = K_5(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + K_6\delta_{ij}\delta_{kl} ,$$

$$\int_0^\infty ds \int d\mathbf{r}'\, \langle [e^{sL}\hat{\tau}_{ij}(0)]\,\hat{\mathcal{T}}_{kl}\, G'_m(\mathbf{r}') r'_n = K_7\delta_{ij}\delta_{kl}\delta_{mn} + K_8\delta_{ij}(\delta_{km}\delta_{ln} + \delta_{kn}\delta_{lm}) + K_9\delta_{kl}(\delta_{im}\delta_{jn} + \delta_{in}\delta_{jm}) + K_{10}\delta_{mn}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})$$
$$+ K_{11}[\delta_{km}(\delta_{il}\delta_{jn} + \delta_{jl}\delta_{in}) + \delta_{lm}(\delta_{ik}\delta_{jn} + \delta_{jk}\delta_{in})] + K_{12}[\delta_{kn}(\delta_{il}\delta_{jm} + \delta_{jl}\delta_{im}) + \delta_{ln}(\delta_{ik}\delta_{jm} + \delta_{jk}\delta_{im})] ,$$

$$\int_0^\infty ds \int d\mathbf{r}'\, \langle [e^{sL}\hat{J}_i^E(0)]\hat{\mathcal{J}}_j^E\, G'_k(\mathbf{r}')\rangle r'_l = K_{13}\delta_{ij}\delta_{kl} + K_{14}\delta_{ik}\delta_{jl} + K_{15}\delta_{il}\delta_{jk} ,$$

$$\int_0^\infty ds \int d\mathbf{r}'\, \langle [e^{sL}\hat{J}_i^E(0)]\,\hat{\mathcal{T}}_{jk} N'(\mathbf{r}')\rangle r'_l = K_{16}(\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl}) + K_{17}\delta_{il}\delta_{jk} ,$$

$$\int_0^\infty ds \int d\mathbf{r}'\, \langle [e^{sL}\hat{J}_i^E(0)]\,\hat{\mathcal{T}}_{jk} E'(\mathbf{r}')\rangle r'_l = K_{18}(\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl}) + K_{19}\delta_{il}\delta_{jk} ,$$

$$\int_0^\infty ds \int d\mathbf{r}'\, \langle [e^{sL}\hat{\tau}_{ij}(0)]\, G_k(\mathbf{r}')\rangle r'_l = K_{20}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + K_{21}\delta_{ij}\delta_{kl} ,$$

$$\int_0^\infty ds \int d\mathbf{r}'\, \langle [e^{sL}\hat{J}_i^E(0)]\, N(\mathbf{r}')\rangle r'_j = K_{22}\delta_{ij} ,$$

$$\int_0^\infty ds \int d\mathbf{r}'\, \langle [e^{sL}\hat{J}_i^E(0)]\, E(\mathbf{r}')\rangle r'_j = K_{23}\delta_{ij} . \tag{A7}$$

The correlation functions in these relations are evaluated in the equilibrium state defined by the value of the thermodynamic variables at point $\mathbf{r}$ and at time t, but in a frame of reference in which $\mathbf{u}(\mathbf{r}, t) = 0$. These correlation functions involve a number of dynamical variables that are defined in the following way. By $N(\mathbf{r})$, $\mathbf{G}(\mathbf{r})$ and $E(\mathbf{r})$ we have denoted the number of particles density, the momentum density and the total energy density:

$$N(\mathbf{r}) = \sum_{\alpha=1}^{N} \delta(\mathbf{r} - \mathbf{r}_\alpha) ,$$

$$\mathbf{G}(\mathbf{r}) = \sum_{\alpha=1}^{N} m\, \mathbf{v}_\alpha\, \delta(\mathbf{r} - \mathbf{r}_\alpha) ,$$

$$E(\mathbf{r}) = \sum_{\alpha=1}^{N} E^{(\alpha)}\, \delta(\mathbf{r} - \mathbf{r}_\alpha) . \tag{A8}$$

Here $\mathbf{r}_\alpha$ and $\mathbf{v}_\alpha$ denote the position and velocity of particle α, N in the number of particles of mass m, and $E^{(\alpha)}$ is the energy of the αth particle

$$E^{(\alpha)} = \tfrac{1}{2} m v_\alpha^2 + \frac{1}{2}\sum_{\beta(\neq\alpha)} V(r_{\alpha\beta}) , \tag{A9}$$

where $r_{\alpha\beta} = |\mathbf{r}_\alpha - \mathbf{r}_\beta|$ and V is the pairwise interaction potential. From the microscopic equations of motion of the density variables (A8) we introduce the local microscopic energy flux $\mathbf{J}^E(\mathbf{r})$ and the momentum flux $\boldsymbol{\tau}(\mathbf{r})$ defined via their Fourier transforms as

$$\mathbf{J}^E(\mathbf{k}) = \sum_{\alpha=1}^{N} [\mathbf{v}_\alpha E^{(\alpha)} + \boldsymbol{\tau}_\alpha(\mathbf{k}) \cdot \mathbf{v}_\alpha] \exp(i\mathbf{k} \cdot \mathbf{r}_\alpha) ,$$

$$\boldsymbol{\tau}(\mathbf{k}) = \sum_{\alpha=1}^{N} [m\mathbf{v}_\alpha\mathbf{v}_\alpha + \boldsymbol{\tau}_\alpha(\mathbf{k})] \exp(i\mathbf{k} \cdot \mathbf{r}_\alpha) ,$$

$$\boldsymbol{\tau}_\alpha(\mathbf{k}) = -\frac{1}{2}\sum_{\beta(\neq\alpha)} \mathbf{r}_{\alpha\beta} \frac{\partial V(r_{\alpha\beta})}{\partial \mathbf{r}_\alpha} \frac{1 - \exp(i\mathbf{k} \cdot \mathbf{r}_{\beta\alpha})}{i\mathbf{k} \cdot \mathbf{r}_{\alpha\beta}} , \tag{A10}$$

where the Fourier transform is defined by

$$f(\mathbf{k})=\int d\mathbf{r}\exp(i\mathbf{k}\cdot\mathbf{r})f(\mathbf{r}) . \tag{A11}$$

The additional phase functions $\hat{J}_i^E(\mathbf{r})$ and $\hat{\tau}_{ij}(\mathbf{r})$ in Eq. (A7) are the subtracted fluxes whose expressions are

$$\hat{J}_i^E(\mathbf{r})=J_i^E(\mathbf{r})-\frac{h}{mn}G_i(\mathbf{r}) ,$$

$$\hat{\tau}_{ij}(\mathbf{r})=\tau_{ij}(\mathbf{r})-\delta_{ij}\left[p+\left(\frac{\partial p}{\partial n}\right)_e N'(\mathbf{r})+\left(\frac{\partial p}{\partial e}\right)_n E'(\mathbf{r})\right] . \tag{A12}$$

The prime on a phase function denotes its deviation from the (reference) equilibrium average, i.e.,

$$G'=G-\langle G\rangle \tag{A13}$$

for any dynamical variable. Also we use the notation

$$\mathcal{J}^E=\int d\mathbf{r}'\,\mathbf{J}^E(\mathbf{r}') ,$$

$$\mathcal{T}_{ij}=\int d\mathbf{r}'\,\tau_{ij}(\mathbf{r}') , \tag{A14}$$

and so on.

To Navier–Stokes order, the shear viscosity η, the bulk viscosity ζ, and the thermal conductivity λ are enough to describe the hydrodynamic behavior of a fluid. Of course, it is easily seen that the expressions reported here for those transport coefficients are the usual Green–Kubo formula.

APPENDIX B: COMPARISON WITH ERNST AND DORFMAN RESULTS

In order to compare the results for the linear Burnett coefficients given in Eqs. (54) and in Ref. 7, we write the linear Burnett transport equations in Fourier space taking, in addition, $\mathbf{k}=k\hat{\mathbf{x}}$ where $\hat{\mathbf{x}}$ is a unit vector in the direction for the x axis. We have, using Eq. (1):

$$\frac{\partial}{\partial t}n(k,t)=-ikn u_x(k,t) ,$$

$$mn\frac{\partial}{\partial t}u_z(k,t)=-ik\langle\tau_{zx}(k)\rangle_t^{(D)} ,$$

$$mn\frac{\partial}{\partial t}u_x(k,t)=-ikp(k,t)-ik\langle\tau_{xx}(k)\rangle_t^{(D)} ,$$

$$\frac{\partial}{\partial t}e(k,t)=-ik(e+p)u_x(k,t)-ik\langle J_x^E(k)\rangle_t^{(D)} , \tag{B1}$$

where

$$\langle\tau_{zx}(k)\rangle_t^{(D)}=-ik\eta u_z(k,t) ,$$

$$\langle\tau_{xx}(k)\rangle_t^{(D)}=ik(\tfrac{2}{3}\eta-\zeta)u_x(k,t)+(ik)^2(\eta_3+\eta_4)T(k,t)-(ik)^2\left(\frac{2\eta_1+\eta_2}{mn}\right)p(k,t) ,$$

$$\langle J_x^E(k)\rangle_t^{(D)}=-ik\lambda T(k,t)+(ik)^2\times\left[\lambda_2-\lambda_1T\left(\frac{\partial p}{\partial e}\right)_n+\lambda_3\right]u_x(k,t) . \tag{B2}$$

We notice that the Fourier transform has been taken considering a near equilibrium situation in which, e.g., the transport coefficients are not functions of the position $\mathbf{r}$. We know that, in fact, they depend on the position through the values of the thermodynamic fields. When these equations are compared with Eqs. (136) and (142) in Ref. 7 we see that they have the same structure and that the correspondence between transport coefficients is the following:

Here	Ernst and Dorfman
λ	λ
$\lambda_2-\lambda_1T\left(\frac{\partial p}{\partial e}\right)_n+\lambda_3$	$-\frac{\gamma-1}{\alpha}O_2+\frac{2}{3}O_4$
η	η
$\frac{2}{3}\eta-\zeta$	$-D_l$
$-\frac{2\eta_1+\eta_2}{mn}$	$-\frac{2}{3}\frac{1}{mn}w_2$
$\eta_3+\eta_4$	$\frac{2}{3}w_3$

(B3)

Also, from Eq. (A6) it follows $(\lambda_2+\lambda_3)/T=\eta_3+\eta_4$ that is equivalent to their relation $w_3=\vartheta_4/T$.

When the low density limit of Eqs. (152) in Ref. 7 is taken and Table (B3) is used, one finds agreement between both theories. (Only the three-dimensional case is considered by Ernst and Dorfman.)

APPENDIX C: LOW-DENSITY LIMIT OF THE CORRELATION FUNCTION $\mu(\tau)$

As a simple example of application of the Eqs. (61), we consider a low density gas. In this approximation, and for relative distances large compared to σ the binary collision operator $\overline{T}_-$ can be replaced by the point operator $T^{(0)}$. We also can drop the last term on the right-hand side of Eq. (61a), so that we find the following equation for $\chi_{1/2}(y_1;\tau/z_1,z_2)$:

$$[\partial_\tau+L_1^0-\lambda^0(1)]\chi_{1/2}(y_1;\tau/z_1,z_2)=\lambda^0(12)\chi_{1/1}(y_1;\tau/z_1)\chi_{1/1}(y_2;\tau/z_2) , \tag{C1}$$

where $\lambda^0(12)$ is given by Eq. (62) but replacing $\overline{T}_-(12)$ by $T^{(0)}(12)$. It may be asked why we keep the term on the right-hand side of Eq. (C1) but we drop the one containing $\chi_{2/2}$ in Eq. (61a), given that both of them seem to involve the same number of particles. Nevertheless, an analysis of Eq. (61b) shows that to lowest order in the density $\chi_{2/2}(\tau)$ gives contributions to the evolution of $\chi_{1/2}(\tau)$ of the form

$$\int dy_2\,T^{(0)}(12)\exp[-\tau(L_1^0+L_2^0)]\chi_{2/2}(y_1,y_2;0/z_1,z_2).$$

Now, the expression of $\chi_{2/2}(0)$ contains equilibrium correlations between particles y_1 and y_2 that are of higher order in density than $\chi_{1/1}(y_1;\tau/z_1)\chi_{1/1}(y_2;\tau/z_2)$.

Let us define the operator

$$\mathfrak{U}(\alpha;\tau)=\exp\{-\tau[L_\alpha^0-\lambda^0(\alpha)]\} . \tag{C2}$$

The formal solution of Eq. (C1) can be expressed as

$$\chi_{1/2}(y_1;\tau/z_1,z_2)=\mathfrak{U}(1;\tau)\chi_{1/2}(y_1;0/z_1,z_2)+\int_0^\tau d\tau_1\,\mathfrak{U}(1;\tau-\tau_1)\lambda^0(12)\chi_{1/1}(y_1;\tau/z_1)\chi_{1/1}(y_2;\tau/z_2) . \tag{C3}$$

The initial term $\chi_{1/2}(0)$ is given in Eq. (63) from where it follows that it can be neglected in the Boltzmann approximation. On the other hand, $\chi_{1/1}$ is to be considered to lowest order in density, i.e., it is given by Eq. (31) or

$$\chi_{1/1}(y_1;\tau_1/z_1)=\exp\{-\tau_1[L_1^0-\lambda^0(1)]\}\delta(y_1-z_1)f_{1,\mathrm{eq}}(y_1)$$
$$=\mathfrak{U}(1;\tau_1)\delta(y_1-z_1)f_{1,\mathrm{eq}}(y_1)\,. \tag{C4}$$

So, for a low-density gas

$$\chi_{1/2}(y_1;\tau/z_1,z_2)=\int_0^\tau d\tau_1\,\mathfrak{U}(1;\tau-\tau_1)\lambda^0(12)[\mathfrak{U}(1;\tau_1)f_{1,\mathrm{eq}}(y_1)$$
$$\times\delta(y_1-z_1)][\mathfrak{U}(2;\tau_2)f_{1,\mathrm{eq}}(y_2)\delta(y_2-z_2)]\,. \tag{C5}$$

From Eqs. (60), (C4), and (C5) it is found

$$\mu(\tau)=\int dy_1\,F_1(y_1)\mathfrak{U}(1;\tau)F_1'(y_1)F_1''(y_1)f_{1,\mathrm{eq}}(y_1)$$
$$+\int_0^\tau d\tau_1\int dy_1\,F_1(y_1)\mathfrak{U}(1;\tau-\tau_1)\lambda^0(12)$$
$$\times[\mathfrak{U}(1;\tau_1)f_{1,\mathrm{eq}}(y_1)F_1'(y_1)][\mathfrak{U}(2;\tau_1)f_{1,\mathrm{eq}}(y_2)F_1''(y_2)]\,. \tag{C6}$$

For a comparison with the expressions obtained by Dufty and used in Refs. 8 and 19 we introduce the operators

$$I(1)\phi(y_1)=\int dy_2 f_{1,\mathrm{eq}}(y_2)T^{(0)}(12)(1+P_{12})\phi(y_1) \tag{C7}$$

and

$$J(12)\varphi(y_1,y_2)$$
$$=\int dy_2 f_{1,\mathrm{eq}}(y_2)T^{(0)}(12)(1+P_{12})\,\varphi(y_1,y_2)\,, \tag{C8}$$

where ϕ and φ are arbitrary functions. Then, Eq. (C6) is equivalent to

$$\mu(\tau)=\int dy_1 f_{1,\mathrm{eq}}(y_1)F_1(y_1)S(1;\tau)F_1'(y_1)F_1''(y_1)$$
$$+\int_0^\tau d\tau_1\int dy_1 f_{1,\mathrm{eq}}(y_1)F_1(y_1)S(1;\tau-\tau_1)J(12)$$
$$\times[S(1;\tau_1)F_1'(y_1)][S(2;\tau_1)F_1''(y_2)]\,, \tag{C9}$$

where

$$S(1;\tau)=\exp\{-\tau[L_1^0+I(1)]\}\,. \tag{C10}$$

The expression (C9) is identical with that used before by Dufty and collaborators. [Compare for instance with Eq. (3.1) in Ref. 8. A difference in sign in the definitions of I and J must be noticed.] It must be emphasized that explicit calculations show that the second addend on the right-hand side of Eq. (C9) gives contributions of the same order as the first one.

APPENDIX D: LONG TIME CONTRIBUTIONS FROM $\chi_{1/2}^{(4)}$ AND $\chi_{1/2}^{(5)}$

In Eq. (68) we replace $\overline{T}_-$ operators by $T^{(0)}$ operators, we use Eq. (72) and introduce the Fourier representation of the δ functions involving position vectors. Then, we proyect onto the hydrodynamic modes and obtain

$$\begin{aligned}\tilde{\chi}_{1/2}^{(4)}(y_1;\epsilon/z_1,z_2)=&\frac{1}{(2\pi)^{3d}}\int d\mathbf{k}_1\int d\mathbf{k}_2\int d\mathbf{k}_3\exp[i(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{r}_1][\epsilon+i(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{v}_1-\lambda_0(1)]^{-1}\int d\mathbf{p}_3\,T_0(13)\frac{1}{n^2}\\&\times{\sum_{\Omega}}'{\sum_{\Omega'}}'[\epsilon+\Omega(l)+\Omega'(k_3)]^{-1}\Theta_0^{(\Omega)}(\hat{\mathbf{l}},\mathbf{v}_1)\Theta_0^{(\Omega')}(-\mathbf{k}_3,\mathbf{v}_3)f_{1,\mathrm{eq}}(y_1)f_{1,\mathrm{eq}}(y_3)\Big\{\int d\mathbf{p}_1\int d\mathbf{p}_3\,\Theta_0^{(\Omega)}(\hat{\mathbf{l}},\mathbf{v}_1)\\&\times\Theta_0^{(\Omega')}(-\mathbf{k}_3,\mathbf{v}_3)T_0(13)(1+P_{13})f_{1,\mathrm{eq}}(y_3)[\epsilon+i(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{v}_1-\lambda_0(1)]^{-1}\exp(-i\mathbf{k}_1\cdot\mathbf{r}_1')\\&\times\exp(-i\mathbf{k}_2\cdot\mathbf{r}_2')\lambda_0(12)[\epsilon+i\mathbf{k}_1\cdot\mathbf{v}_1+i\mathbf{k}_2\cdot\mathbf{v}_2-\Lambda_0(12)]^{-1}\delta(\mathbf{p}_1-\mathbf{p}_1')\delta(\mathbf{p}_2-\mathbf{p}_2')f_{1,\mathrm{eq}}(y_1)f_{1,\mathrm{eq}}(y_2)\Big\}\,.\end{aligned} \tag{D1}$$

Here $\mathbf{l}=\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3$ and we have used the notation $y_1=\{\mathbf{r}_1,\mathbf{p}_1\}$ and $z_\alpha=\{\mathbf{r}_\alpha',\mathbf{p}_\alpha'\}$, $\alpha=1,2$. Now we consider the two cases that are interesting in the calculation of the nonlinear Burnett transport coefficients

$$\text{(I)}\quad F_1(y_1)=F_1(\mathbf{p}_1)\,,\quad F_1'(z_1)=F_1'(\mathbf{p}_1')\,,\quad F_1''(z_2)=F_1''(\mathbf{p}_2') \tag{D2}$$

and

$$\text{(II)}\quad F_1(y_1)=F_1(\mathbf{p}_1)\delta(\mathbf{r}_1)\,,\quad F_1'(z_1)=F_1'(\mathbf{p}_1')\,,\quad F_1''(z_2)=F_1''(\mathbf{p}_2')\mathbf{r}_2'\,. \tag{D3}$$

Then, we introduce Eq. (D1) in the first term on the right-hand side of Eq. (60) and evaluate the long time contributions to $\mu(\tau)$ in both cases (I) and (II). After some manipulations and using the same properties already discussed in Sec. III it is found

$$\mu_{\mathrm{I}}^{(4)}(\tau)=\frac{V}{2n^2}{\sum_{\Omega}}'{\sum_{\Omega'}}'\int\frac{d\mathbf{k}}{(2\pi)^d}\exp\{-\tau[\Omega(k)+\Omega'(k)]\}I_{\Omega,\Omega'}(\hat{\mathbf{k}},F_1)K_{\Omega,\Omega'}(\hat{\mathbf{k}},F_1',F_1'')\,, \tag{D4}$$

$$\mu_{\mathrm{II}}^{(4)}(\tau)=-\frac{1}{n^2}\tau\int\frac{d\mathbf{k}}{(2\pi)^d}c_0\hat{\mathbf{k}}\exp(-\tau\Gamma_{s,0}k^2)I_{+,-}(\hat{\mathbf{k}},F_1)K_{+,-}(\hat{\mathbf{k}},F_1',F_1'')\,, \tag{D5}$$

where $I_{\Omega,\Omega'}$ is given by Eq. (48) and

$$K_{\Omega,\Omega'}(\hat{\mathbf{k}},F_1',F_1'')=\int d\mathbf{p}_1\,\Theta_0^{(\Omega)}(\hat{\mathbf{k}},\mathbf{v}_1)\Theta_0^{(\Omega')}(-\hat{\mathbf{k}},\mathbf{v}_1)\lambda_0(12)[-\Lambda_0(12)]^{-1}F_1'(\mathbf{p}_1)F_1''(\mathbf{p}_2)f_{1,\mathrm{eq}}(y_1)f_{1,\mathrm{eq}}(y_2)\,. \tag{D6}$$

We notice that the evaluation of $K_{\Omega,\Omega'}$ is not trivial. One could use as a first estimate the first Enskog approximation.

The calculations for $\chi_{1/2}^{(5)}$ and then for $\mu_{I}^{(5)}(\tau)$ and $\mu_{II}^{(5)}(\tau)$ are quite similar. The final result is that

$$\mu_{I}^{(5)}(\tau) = -\mu_{I}^{(4)}(\tau), \quad \mu_{II}^{(5)}(\tau) = -\mu_{II}^{(4)}(\tau) \tag{D7}$$

thus cancelling each other. The difference in sign is due, from a mathematical point of view, to the extra operator $[\epsilon + L_1^0 - \lambda(1)]^{-1}$ contained in Eq. (68).

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