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A Convergent Nonequilibrium Statistical Mechanical Theory for Dense Gases. I. The Two-Body Distribution Function

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We analyze the kinetic theory for dense gases proposed by Bogolyubov. We arrive at the conclusion that the boundary conditions proposed by him are not adequate for the description of a dense gas. This fact is the reason for the occurrence of the divergences found in the virial expansion of the generalized Boltzmann equation and of the transport coefficients. We propose new boundary conditions which lead to convergent virial expansions of the kinetic equation and of the transport coefficients.

KEY WORDS: Convergent kinetic theory; dense gases; boundary conditions; divergences in the transport coefficients; convergent virial expansion; two-body distribution function.

1. INTRODUCTION

In 1946 Bogolyubov[1] presented his theory of nonequilibrium statistical mechanics for dense gases. In the development of this theory three essential assumptions are made: (i) the functional hypothesis, (ii) the existence of a

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density expansion of the relevant quantities, and (iii) certain boundary conditions in order to solve the BBGKY hierarchy.

Since then much work has been invested in obtaining all its possible consequences.[2] In particular, attention was centered on the explicit derivation of a virial expansion of the kinetic equation that describes the evolution of the system, and of the corresponding transport coefficients. In this way an attempt was made to generalize the Boltzmann equation to higher orders in the density. However, when calculations were made to obtain the second order in the density of the transport coefficients, it was found that the resulting expressions diverged.[3] This means that the configuration integrals involved diverge. For higher orders in the density, the same sort of divergences were obtained. Thereafter the idea of a virial expansion of the transport coefficients was severely questioned. In order to remedy this situation, several methods have been proposed. We only mention the resummation technique followed by Dorfman *et al.*[4]

On the other hand, methods have been developed for calculating linear transport coefficients without any reference to a density expansion.[5] However, these developments are formal, in the sense that they still depend on the two-body distribution, which so far is not available without a density expansion.

It should be mentioned that there have also been workers who have implied that no functional description in terms of the single-particle distribution function is possible. This means that no kinetic equation for the system exists.

However, the use of the hypotheses concerning the nonexistence of the virial expansion and/or the functionality has not led to satisfactory results.

The third hypothesis used by Bogolyubov, the one concerning the boundary conditions, has, to our knowledge, not been analyzed earlier by other authors.[6]

It is the purpose of this paper to analyze the boundary conditions introduced by Bogolyubov. We will show that these boundary conditions are not adequate for the description of a dense gas. The appearance of the divergences is shown to be a consequence of not taking the medium into account in the boundary conditions. We will propose new boundary conditions that do take into account the medium explicitly, and which lead to convergent virial expansions of the kinetic equation and of the transport coefficients.

The appearance of the above-mentioned divergences has already been doubted by several authors.[7] We only mention the work of Fujita[8] using diagrammatic methods and of Byung Chan Eu[9] starting from the method of correlation functions.

In this paper we obtain the first terms in the density expansion of the two-body distribution function. In forthcoming papers we will present the calculations of the different terms in the density expansion of the transport coefficients.

In Section 2 we solve formally the BBGKY hierarchy using only two assumptions: (i) the functional hypothesis and (ii) the existence of a density expansion. In Section 3 we discuss Bogolyubov's boundary conditions and show that they are not adequate. We then proceed to propose new boundary conditions. In Section 4 we use the new boundary conditions in order to obtain explicitly the two-body distribution function as functional of the single distribution function, to different orders in the density. Finally in Section 5 we sum up the results, and conclude that there exists a density expansion of the kinetic equation and of the transport coefficients to any order in the density.

2. FORMAL SOLUTION OF THE BBGKY HIERARCHY

Let us consider a one-component gas consisting of N molecules of mass m enclosed in a volume V. The Hamiltonian of this system will be taken of the form

$$H = \sum_{i=1}^{N} (p_i^2/2m) + \tfrac{1}{2} \sum\sum_{i \neq j}^{N} \varphi(|\,\mathbf{q}_i - \mathbf{q}_j\,|) \tag{1}$$

where $\mathbf{p}_i$ and $\mathbf{q}_i$ are the momentum and position of the ith particle, respectively. Here φ is the pair potential, which we restrict to be a strong repulsive potential.

As is well known, the BBGKY hierarchy can be written as follows:

$$(\partial F_s/\partial t) + \mathscr{H}_s F_s = n \int dx_{s+1} \sum_{i=1}^{s} \theta_{i,s+1} F_{s+1}\,, \qquad s = 1, 2, \ldots \tag{2}$$

Here we have already taken the so-called thermodynamic limit, $N \to \infty$, $V \to \infty$, $N/V = n$. In this expression $F_s(x_1, \ldots, x_s\,; t)$ denotes the s-body distribution function in phase space, and $x_i \equiv (\mathbf{p}_i\,, \mathbf{q}_i)$. The operators θ_{ij} and $\mathscr{H}_s$ are given by

$$\theta_{ij} = \frac{\partial\varphi(|\,\mathbf{q}_i - \mathbf{q}_j\,|)}{\partial \mathbf{q}_i} \cdot \frac{\partial}{\partial \mathbf{p}_i} + \frac{\partial\varphi(|\,\mathbf{q}_i - \mathbf{q}_j\,|)}{\partial \mathbf{q}_j} \cdot \frac{\partial}{\partial \mathbf{p}_j} \tag{3}$$

and

$$\mathscr{H}_s = \sum_{i=1}^{s} \frac{\mathbf{p}_i}{m} \cdot \frac{\partial}{\partial \mathbf{q}_i} - \sum\sum_{i<j}^{s} \theta_{ij} \tag{4}$$

As was mentioned in the introduction, we will assume that in the kinetic stage of the evolution of the system the s-body distribution function ($s \geqslant 2$) is a time-independent functional of F_1, i.e.,

$$F_s(x_1, \ldots, x_s; t) \rightarrow F_s(x_1, \ldots, x_s \mid F_1) \tag{5}$$

where the usual notation is used. Substituting Eq. (5) into the first equation of the BBGKY hierarchy ($s = 1$), one finds the kinetic equation, valid to any order in the density,

$$\partial F_1/\partial t = A(x_1 \mid F_1) \tag{6}$$

Let us expand A and F_s in powers of the density:

$$A(x_1 \mid F_1) = \sum_{l=0}^{\infty} n^l A^{(l)}(x_1 \mid F_1) \tag{7}$$

and

$$F_s(x_1, \ldots, x_s \mid F_1) = \sum_{l=0}^{\infty} n^l F_s^{(l)}(x_1, \ldots, x_s \mid F_1) \tag{8}$$

Substituting Eq. (7) into Eq. (6), and Eq. (8) for $s = 1$ into the first equation of the hierarchy, and comparing the resulting expressions, one finds that

$$A^{(0)}(x_1 \mid F_1) = -(\mathbf{p}_1/m) \cdot (\partial F_1/\partial \mathbf{q}_1) \tag{9}$$

$$A^{(l)}(x_1 \mid F_1) = \int dx_2\, \theta_{12} F_2^{(l-1)}(x_1, x_2 \mid F_1), \qquad l \geqslant 1 \tag{10}$$

Proceeding as usual, we write

$$\partial F_s(x_1, \ldots, x_s \mid F_1)/\partial t = [\delta F_s/\delta F_1, \partial F_1/\partial t] = [\delta F_s/\delta F_1, A(x_1 \mid F_1)] \tag{11}$$

where $(\delta F_s/\delta F_1)$ denotes the functional derivative of F_s with respect to F_1. Substituting Eq. (11) into the hierarchy, Eq. (2), we find the following differential equations, to the various orders in the density:

$$D^{(0)} F_s^{(0)} + \mathscr{H}_s F_s^{(0)} = 0 \tag{12}$$

$$\begin{aligned} D^{(0)} F_s^{(l)} + \mathscr{H}_s F_s^{(l)} &= -\sum_{j=1}^{l} D^{(j)} F_s^{(l-j)} + \int dx_{s+1} \sum_{i=1}^{s} \theta_{i,s+1} F_{s+1}^{(l-1)} \\ &\equiv \psi_s^{(l)}(x_1, \ldots, x_s \mid F_1), \qquad l \geqslant 1 \end{aligned} \tag{13}$$

Here the operators $D^{(k)}$ are defined as

$$D^{(k)}P_s(x_1, ..., x_s \mid F_1) = [\delta P_s/\delta F_1, A^{(k)}(x_1 \mid F_1)] \tag{14}$$

As is well known,[1] the solutions of Eqs. (12) and (13) are

$$F_s^{(0)}(x_1, ..., x_s \mid F_1) = S_s^{-\tau} F_s^{(0)}(x_1, ..., x_s \mid S_1^{\tau} F_1) \tag{15}$$

and

$$F_s^{(l)}(x_1, ..., x_s \mid F_1) = S_s^{-\tau} F_s^{(l)}(x_1, ..., x_s \mid S_1^{\tau} F_1) + \int_0^{\tau} d\tau\, S_s^{-\tau} \psi_s^{(l)}(x_1, ..., x_s \mid S_1^{\tau} F_1), \qquad l \geqslant 1 \tag{16}$$

The streaming operator $S_s^{\tau}(x_1, ..., x_s)$ of s particles is given by

$$S_s^{\tau}(x_1, ..., x_s) = \exp[\tau \mathscr{H}_s(x_1, ..., x_s)] \tag{17}$$

It should be mentioned that these results are obtained by making only two assumptions: (i) the functional hypothesis [Eq. (5)], and (ii) the existence of a density expansion.

It is at this point where various authors use the boundary conditions advanced by Bogolyubov, in order to obtain explicitly the functional dependence of F_s [Eq. (5)]. In the following section we will discuss new boundary conditions that take into account the medium in the hydrodynamic stage.

3. BOUNDARY CONDITIONS

First of all we will discuss the physical meaning of Bogolyubov's boundary conditions (BBC). These are given by

$$\lim_{\tau \to \infty} S_s^{-\tau} F_s(x_1, ..., x_s \mid S_1^{\tau} F_1) = \lim_{\tau \to \infty} S_s^{-\tau} \prod_{i=1}^{s} S_1^{\tau} F_1(x_i ; t) \tag{18}$$

The physical meaning of BBC is that when several particles are out of their mutual sphere of influence (the range of the potential) then there are no correlations between them. The only correlations allowed for by BBC will occur only once, namely when these several particles collide. This is an attempt to generalize the Stosszahlansatz introduced by Boltzmann for dilute gases. However, the BBC are not correct for the case of a dense gas.

In fact, let us consider the case of a gas of hard spheres in equilibrium. When they are in contact then they are, of course, correlated. As is well known, this correlation persists even when the spheres are out of their mutual sphere of influence. This correlation is atributed to an effective mean force between the two spheres *through* the medium in which they are embedded. This medium consists of the remaining particles of the system. This same property will be manifested even if the system is in a nonequilibrium state.

It should be noted that this influence of the medium will be manifested in a stronger way as the density increases. For the case of the dilute gas one should recover the Stosszahlansatz hypothesis.

We now see that the boundary conditions given by Eq. (18) do not exhibit this property. This fact, the absence of the medium in the boundary conditions given by Bogolyubov, is precisely the reason of the divergences that have appeared.[3] The only correlations taken into account in Eq. (18) are collisional ones due to direct forces between the particles.

Now we proceed to propose boundary conditions that take into account the influence of the medium. For this purpose, we note that the contribution due to the *direct* forces between the particles can be expressed, to zeroth order in the density, as follows[10]:

$$\lim_{\tau\to\infty} S_s^{-\tau} F_s^{(0)}(x_1, \ldots, x_s \mid S_1^{\tau} F_1) = \lim_{\tau\to\infty} S_s^{-\tau} \prod_{i=1}^{s} S_1^{\tau} F_1(x_i ; t) \tag{19a}$$

and to higher orders in the density as

$$\lim_{\tau\to\infty} S_s^{-\tau} F_s^{(l)}(x_1, \ldots, x_s \mid S_1^{\tau} F_1) = 0, \qquad l \neq 0 \tag{19b}$$

If the medium is taken into account, one should write, to zeroth order in the density, instead of Eq. (19a),

$$\lim_{\tau\to\infty} S_s^{-\tau} F_s^{(0)}(x_1, \ldots, x_s \mid S_1^{\tau} F_1) = (1 - g_s^{(0)}) \lim_{\tau\to\infty} S_s^{-\tau} \prod_{i=1}^{s} S_1^{\tau} F_1(x_i ; t) \tag{20a}$$

and to higher orders in the density, instead of Eq. (19b),

$$\lim_{\tau\to\infty} S_s^{-\tau} F_s^{(l)}(x_1, \ldots, x_s \mid S_1^{\tau} F_1) = g_s^{(l)} \lim_{\tau\to\infty} S_s^{-\tau} \prod_{i=1}^{s} S_1^{\tau} F_1(x_i ; t), \qquad l \neq 0 \tag{20b}$$

Here g_s is the local equilibrium correlation function between s-particles, and is expressed as a power series in the density

$$g_s = \sum_{l=0}^{\infty} n^l g_s^{(l)} \tag{21}$$

We have used the local equilibrium correlation function because we want to calculate properties of our system in the hydrodynamic stage. At this point we would like to emphasize the fact that boundary conditions in general, and in particular either BBC or our new boundary conditions, are hypotheses which are not derivable from the very formalism of the theory. In other words, boundary conditions are assumptions which one makes about the behavior of the system, and which one introduces in order to solve the differential equations describing the general laws that the system obeys.

Combining Eqs. (20) and (21), one can write the boundary conditions compactly as

$$\lim_{\tau\to\infty} S_s^{-\tau} F_1(x_1 ,\dots, x_s \mid S_1{}^\tau F_1) = (1 - 2g_s^{(0)} + g_s) \lim_{\tau\to\infty} S_s^{-\tau} \prod_{i=1}^{s} S_1{}^\tau F_1(x_i\,;t) \tag{22}$$

The physical meaning of Eq. (22) is that a set of particles that collide once will be allowed to collide many more times because the probability that they will be far apart *after the collision* will be very small. This is a consequence of the effect of the medium; the medium does not allow the interacting particles to get far apart. In other words, the medium will, on the average, confine the interacting set of particles to some region in space. The dimensions of this region will, of course, depend on the potential and on the density.

We can also conclude from the boundary conditions (22) that particles that are too far away from each other will have a very small probability of colliding, and therefore they are independent. This is again an effect of the medium.

It should be mentioned that BBC only take into account dynamic events, whereas our new boundary conditions take also statistical effects into account.

In the next section we will use the new boundary conditions (22) and obtain explicitly the functional dependence of the distribution function F_2 to several orders in the density.

4. THE TWO-BODY DISTRIBUTION FUNCTION

In this section we will obtain the two-body distribution function to first order in the density. The other distribution functions F_s $(s > 2)$ can be obtained in an analogous way.

To zeroth order in the density, we substitute Eq. (20a) into Eq. (15) and find

$$F_s^{(0)}(x_1 ,\dots, x_s \mid F_1) = \Gamma_s(\mathbf{q}_1 ,\dots, \mathbf{q}_s)\, \mathscr{S}_s(x_1 ,\dots, x_s) \prod_{i=1}^{s} F_1(x_i\,;t) \tag{23}$$

Here we have written

$$\Gamma_s(\mathbf{q}_1 ,\dots, \mathbf{q}_s) = 1 - g_s^{(0)}(\mathbf{q}_1 ,\dots, \mathbf{q}_s) \tag{24}$$

and

$$\mathscr{S}_s(x_1, ..., x_s) = \lim_{\tau\to\infty} S_s^{-\tau}(x_1, ..., x_s) \prod_{i=1}^{s} S_1^{\tau}(x_i) \tag{25}$$

For $s = 2$ we obtain from Eq. (23) the zeroth-order term in the two-body distribution function as follows:

$$F_2^{(0)}(x_1, x_2 \mid F_1) = \Gamma_2(\mathbf{q}_1, \mathbf{q}_2)\, \mathscr{S}_2(x_1, x_2) \prod_{i=1}^{2} F_1(x_i; t) \tag{26}$$

Starting from Eq. (16) for $s = 2$ and with the help of Eq. (20b) one finds the first-order term in the density of the two-body distribution function

$$F_2^{(1)}(x_1, x_2 \mid F_1) = g_2^{(1)}(\mathbf{q}_1, \mathbf{q}_2)\, \mathscr{S}_2(x_1, x_2) \prod_{i=1}^{2} F_1(x_i; t) + \int_0^\infty d\tau\, S_2^{-\tau}(x_1, x_2)\, \psi_2^{(1)}(x_1, x_2 \mid S_1^{\tau} F_1) \tag{27}$$

Proceeding in the usual way,[2] one obtains from Eqs. (13)–(15)

$$\begin{aligned} F_2^{(1)}&(x_1, x_2 \mid F_1) \\ &= g_2^{(1)}(\mathbf{q}_1, \mathbf{q}_2)\, \mathscr{S}_2(x_1, x_2) \prod_{i=1}^{2} F_1(x_i; t) \\ &+ \int_0^\infty d\tau\, S_2^{-\tau}(x_1, x_2) \int dx_3 \{(\theta_{13} + \theta_{23})\, \Gamma_3(\mathbf{q}_1, \mathbf{q}_2, \mathbf{q}_3)\, \mathscr{S}_3(x_1, x_2, x_3) \\ &- \Gamma_2(\mathbf{q}_1, \mathbf{q}_2)\, \mathscr{S}_2(x_1, x_2)[\theta_{23}\Gamma_2(\mathbf{q}_2, \mathbf{q}_3)\, \mathscr{S}_2(x_2, x_3) \\ &+ \theta_{13}\Gamma_2(\mathbf{q}_1, \mathbf{q}_3)\, \mathscr{S}_2(x_1, x_3)]\} \prod_{i=1}^{3} F_1(x_i; t) \end{aligned} \tag{28}$$

After a lengthy calculation analogous to the one made by Cohen one finally finds[3] that

$$F_2^{(1)}(x_1, x_2 \mid F_1) = g_2^{(1)}(\mathbf{q}_1, \mathbf{q}_2)\, \mathscr{S}_2(x_1, x_2) \prod_{i=1}^{2} F_1(x_i; t) + \int dx_3\, \mathscr{O}_3(x_1, x_2, x_3) \prod_{i=1}^{3} F_1(x_i; t) \tag{29}$$

[3] The detailed calculations leading to Eq. (30) are available from the authors upon request.

The operator $\mathscr{O}_3(x_1, x_2, x_3)$ is given by

$$\begin{aligned}\mathscr{O}_3(x_1, x_2, x_3) = {} & R(x_1, x_2, x_3) - S_2^{\infty}(x_1, x_2)[\Gamma_2(\mathbf{q}_1, \mathbf{q}_3)\, \mathscr{S}_2(x_1, x_3) \\ & + \Gamma_2(\mathbf{q}_2, \mathbf{q}_3)\, \mathscr{S}_2(x_2, x_3)] \\ & + \int_0^{\infty} d\tau\, S_2^{-\tau}(x_1, x_2)\langle[\hat{h}\Gamma_2(\mathbf{q}_1, \mathbf{q}_2)]\, \mathscr{S}_2(x_1, x_2) \\ & \times [\Gamma_2(\mathbf{q}_2, \mathbf{q}_3)\, \mathscr{S}_2(x_2, x_3) + \Gamma_2(\mathbf{q}_1, \mathbf{q}_3)\, \mathscr{S}_2(x_1, x_3) - 1] \\ & - \{[\hat{h}\Gamma_3(\mathbf{q}_1, \mathbf{q}_2, \mathbf{q}_3)]\, \mathscr{S}_3(x_1, x_2, x_3) \\ & - \Gamma_2(\mathbf{q}_1, \mathbf{q}_2)\, \mathscr{S}_2(x_1, x_2)[(\hat{h}\Gamma_2(\mathbf{q}_2, \mathbf{q}_3))\, \mathscr{S}_2(x_2, x_3) \\ & + (\hat{h}\Gamma_2(\mathbf{q}_1, \mathbf{q}_3))\, \mathscr{S}_2(x_1, x_3)]\}\rangle \end{aligned} \tag{30}$$

Here

$$\begin{aligned} R_3(x_1, x_2, x_3) = {} & \Gamma_3(\mathbf{q}_1, \mathbf{q}_2, \mathbf{q}_3)\, \mathscr{S}_3(x_1, x_2, x_3) \\ & - \Gamma_2(\mathbf{q}_1, \mathbf{q}_2)\, \mathscr{S}_2(x_1, x_2)[\Gamma_2(\mathbf{q}_2, \mathbf{q}_3)\, \mathscr{S}_2(x_2, x_3) \\ & + \Gamma_2(\mathbf{q}_1, \mathbf{q}_3)\, \mathscr{S}_2(x_1, x_3) + \Gamma_2(\mathbf{q}_1, \mathbf{q}_2)\, \mathscr{S}_2(x_1, x_2)] \end{aligned} \tag{31}$$

and

$$\hat{h} = \sum_{i=1}^{3} \mathscr{H}_1(x_i) \tag{32}$$

We have also explicitly obtained $F_2^{(2)}$. However, since the resulting expression is quite long, we will not write it out here.

5. DISCUSSION

In this paper we have started from the usual BBGKY hierarchy and solved it using new boundary conditions. These boundary conditions, as opposed to the ones introduced by Bogolyubov, explicitly take into account the medium.

From inspection of the expressions obtained in Section 4 we now see that if we substitute Eqs. (26) and (29) and the expressions for the higher-order terms of the two-body distribution function into the first equation of the BBGKY hierarchy, the resulting kinetic equation is convergent to any order in the density because the terms $(1 - g_s^{(0)})$ and $g_s^{(l)}$ act as convergence factors in the configurational integrals. Furthermore, one can also show[11] that the corresponding transport coefficients converge to all orders in the density. It is convenient to remark that the dynamic events which produce the divergences in the usual theory are also present in our theory. However, in our case they do not contribute because they are weighted by the factors Γ_s and $g_s^{(l)}$.

Therefore, we can conclude the existence of a virial expansion of the transport coefficients for a dense gas.

Recent accurate measurements on the transport coefficients over a wide range of densities show that the best fit to the experimental data is a power series in the density.[12]

At this point we would like to emphasize that the results obtained in this paper are valid only in the hydrodynamic regime, and we do not pretend to describe the evolution of the system from certain initial conditions. As a matter of fact, the new boundary conditions proposed were presented in order to solve the differential equations satisfied by F_s and to obtain the transport coefficients of the system.

It is worth mentioning that it is conceivable to use other mathematical expressions for boundary conditions which reflect the same physical effects of the medium as discussed in the paragraphs after Eq. (22). All these different ways of expressing the boundary conditions will give rise the same values of the transport coefficients.

The particular choice of our new boundary conditions arose from the need to also include in them statistical effects of the medium, which are described by the correlation function, at least in the hydrodynamic regime. Therefore this function must appear explicitly in the boundary conditions. The simplest way of introducing it is given by Eqs. (20). Of course, the justification of this choice, as of any other choice, has to be made *a posteriori*. That is, when the numerical results obtained from the theory are tested against *real* experimental results. In this sense, the usual Bogolyubov theory has failed. Of course, we do not consider as real experimental data the molecular dynamics results.

In forthcoming papers[11] we will present the explicit calculations of the virial expansion of the transport coefficients.

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