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Discrete Hydrodynamics: Determination of Equations of Motion by Simulation

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A detailed procedure is described for computing discrete equations of motion for a fluid, to implement a new method which turns out to be substantially more efficient than previous methods for calculating transport coefficients. This paper describes the calculations of discrete averages from molecular dynamics data and the numerical extraction of the equation-of-motion coefficients in a way which makes maximum use of the geometric symmetry of the problem. Extrapolation to the infinite-system limit and eventual computation of transport coefficients by renormalization are discussed. The method described in detail here is briefly sketched and applied numerically to computing the viscosity of the soft-sphere liquid in a subsequent paper.

KEY WORDS: Molecular dynamics; transport coefficients; viscosity.

1. INTRODUCTION

Recently a discrete formulation of hydrodynamics was proposed which appears to treat fluid motions in a more natural way than continuum theories; it was shown to be as complete a coarse-grained description of a fluid as possible, in a certain sense.⁽¹⁾ Since then prescriptions have been found for extracting the discrete equations of motion from ensemble averages of discrete variables,⁽²⁾ and for renormalizing the equations of motion to give the macroscopic transport coefficients.⁽³⁾ The purpose of the present paper is to give a detailed and workable method for the computation of the equations of motion from molecular dynamics data on finite systems. An accompanying paper⁽⁴⁾ gives numerical results for the soft-sphere liquid ($\phi \propto r^{-12}$), for which the transport coefficients have been previously calculated⁽⁵⁾; our

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results are consistent with the previous ones, but require about 100 times less calculation.

The discrete hydrodynamic variables and equations of motion have been described in detail previously.$^{(3,2)}$ Briefly, a fluid is subdivided into a simple cubic lattice of cells of width W, whose contents of the conserved quantities momentum, energy, and number (at discrete times $m\tau$, integer multiples of some basic time τ) are the fundamental variables of the theory. Such a content is denoted by $c_{\alpha lm}$, where $\alpha = P_x, P_y, P_z, E$, or N indicates which content is meant, l is a cell label, and m indicates the time. The discrete equation of motion will predict the transfer $x_{\alpha fm}$ of each quantity α across each face (labeled f) during each time interval (labeled by its midpoint m, a half-integer); it is the analog of the continuum equation of motion which predicts fluxes. Details of the numerical calculation of the contents and transfers from molecular dynamics trajectories are given in an accompanying paper.$^{(4)}$

The discrete equation of motion gives the conditional mean transfer $x_{\alpha f1/2}$ over the interval $(0, \tau)$ in terms of the history of the system (contents $c_{\alpha l0}$ and transfers $x_{\alpha fm}$ with $m < 0$). We will denote a compound subscript αfm or αlm by j for brevity, and a history variable by h_j (a content if m is an integer, transfer if not). The conditional mean is parametrized as a power series in the h_j; given a set of indices (some of which may be identical) $\underline{j} = \{j_1, j_2, ...\}$, the product of the corresponding h_j is denoted by $h_{\underline{j}}$. A complete power series is thus

$$[x_{\underline{j}}] = \sum_{\underline{k}} B_{\underline{j};\underline{k}} h_{\underline{k}} \tag{1.1}$$

where we have described conditional moments as well as means by replacing x_j by an arbitrary product $x_{\underline{j}}$. As in Ref. 3, we have subtracted certain nominal values from the contents and transfers so they are small near equilibrium; hence the power series converges rapidly. The coefficients B provide an exactly renormalizable description of the system.$^{(3)}$

The B's can be numerically calculated by doing a molecular dynamics simulation. The direct results of such a simulation are equilibrium averages of products of contents and transfers. Denoting a general discrete variable (content or transfer) by v_j, such an average will be denoted $\langle v_{\underline{j}} \rangle$. The averages are related to the B's through equations obtainable$^{(2)}$ from Eq. (1.1) by multiplying by $h_{\underline{k}'}$ and averaging:

$$\langle x_{\underline{j}} h_{\underline{k}'} \rangle = \sum_{\underline{k}} B_{\underline{j};\underline{k}} \langle h_{\underline{k}} h_{\underline{k}'} \rangle \tag{1.2}$$

Computation of the macroscopic transport coefficients requires renormalization of the B's (i.e., repeatedly doubling W and τ).$^{(3)}$ This is most easily done using an equivalent but slightly different parametrization of the condition mean, in terms of the "excess-transfer parameters" $\tilde{B}_{\underline{j};\underline{k}}$. (These differ only in how they treat the possible non-Markovian dependence of Eq. (1.1) on past transfers x_k; they are an expansion in Δx_k, the excess above the expected value $[x_k]$.) Since we renormalize in an infinite system, we require the infinite-system limit $\tilde{B}^{\infty}_{j;k}$ of the small-cell equations of motion.

In this paper we concern ourselves with determining this $\tilde{B}^{\infty}$ (which can be thought of as giving "bare" transport coefficients) from raw molecular dynamics data (a time series of the v_j). The procedures required are (1) averaging the data to estimate $\langle \underline{v}_j \rangle$, (2) obtaining B's from Eq. (1.2), (3) obtaining $\tilde{B}$ from B, and (4) extrapolating to the infinite system. Obviously step 1 must be done first. In Section 4 a strong argument is given that step 4 (extrapolation) should not precede step 2 because the conditional means (B or $\tilde{B}$) converge much faster than ensemble averages to their infinite-system values. The extrapolation procedure described in Section 4 seems to apply best to $\tilde{B}$, implying that step 3 precedes step 4. Thus we conclude that steps 1–4 should be done in that order; they are described respectively in Section 2, Section 3, Ref. 4, and Section 4.

2. COMPUTING NONEQUIVALENT AVERAGES: SYMMETRY

We wish to determine the equation-of-motion coefficients $B_{j;\underline{k}}$ from Eq. (1.2), which requires estimates $\langle \underline{v}_j \rangle$ of finite-system ensemble averages. These we must obtain from raw molecular dynamics data v_j^{MD} (calculated as described in Ref. 4 and stored on magnetic tape) containing five contents and 15 transfers for each cell and each time interval calculated (i.e., those whose time index m satisfies $0 < m \leqslant m_{\mathrm{tot}}$, if the duration of the run is $m_{\mathrm{tot}}\tau$). Clearly, many of the B's are equivalent because of rotational and translational symmetry, so we wish only to calculate one from each equivalence class. Similarly many $\underline{v}_j$ are equivalent (hence their averages $\langle \underline{v}_j \rangle$ are the same in a symmetry-invariant ensemble such as the microcanonical one used in molecular dynamics). Products of raw data $\underline{v}_j^{\mathrm{MD}}$ are of course not invariant, even after the usual time-averaging; clearly we obtain the "best statistics" by averaging $\underline{v}_j^{\mathrm{MD}}$ over all equivalent variable-products $\underline{v}_j$.

Specifically, the group G under which our system is invariant is the product of the cubic group O_h (Schönflies notation) of rotations about some origin, say a cube corner, and the translation group T of our cell lattice (an infinite group for an infinite lattice, but finite for our finite, periodic molecular dynamics systems. We assume the finite systems are cubical so O_h symmetry may still be used). An operation $g \in G$ acts on cell labels l and face labels f in the obvious way$^{(1)}$; its action can be defined on the compound label j (i.e., αlm or αfm).

The sign change in the content or transfer v_j resulting from g (defined precisely in Section 5 of Ref. 1) is denoted $\rho(j; g)$. Then the invariance of the B coefficients is expressed by

$$B_{\underline{j};\underline{k}} = \rho(\underline{j} \cup \underline{k}; g) B_{g\underline{j};g\underline{k}} \tag{2.1}$$

where

$$\rho(\underline{j}; g) \equiv \prod_{j \in \underline{j}} \rho(j; g) \tag{2.2}$$

Let us label the equivalence classes of combined sets $\underline{j} \cup \underline{k}$ by integers β for convenience in computing; a few such classes are listed in Table I of Ref. 4. Pick some arbitrary representative, denoted $\underline{j}(\beta)$, $\underline{k}(\beta)$ of each class. We need only calculate

$$B_\beta \equiv B_{j(\beta);k(\beta)} \tag{2.3}$$

for each class β.

Enumeration of equivalence classes of products $\underline{v}_{\underline{j}}$ (whose averages differ only in sign: $\langle \underline{v}_{\underline{j}} \rangle = \rho(\underline{j}, g) \langle \underline{v}_{g\underline{j}} \rangle$) is similar. However, the relevant group (call it G') is larger; it includes time translation (by multiples of τ) and time reversal (which introduces into ρ a sign change for each momentum and each transfer) because these are symmetries of the equations of motion. We label the equivalence classes of $\underline{j}$'s by integers γ. These classes are different from those defined for the B's. Because $G \subset G'$, each class β is a subclass of some γ, which we shall call $\gamma(\beta)$; γ may contain several β's. For convenience, we may choose labels so that γ is the same as one of these β's. [In Table II of Ref. 4, $\gamma(\beta) = \beta$, but this will not always be true.] The nonequivalent averages we wish to compute are therefore

$$\langle \underline{v}_\gamma \rangle \equiv \langle \underline{v}_{\underline{j}'(\gamma)} \rangle \tag{2.4}$$

where $\underline{j}'(\gamma)$ is an arbitrary representative of class γ.

These are best estimated from the raw data $v_{\underline{j}}^{\mathrm{MD}}$ by averaging over time, rotations, and translations:

$$\langle \underline{v}_\gamma \rangle = \frac{1}{N} \sum_{g\underline{j}'(\gamma)} \underline{v}^{\mathrm{MD}}_{g\underline{j}'(\gamma)} \tag{2.5}$$

where the sum is over all index sets $g\underline{j}'(\gamma)$ that are equivalent under G' to $\underline{j}'(\gamma)$ and all of whose time indices m satisfy $0 < m \leqslant m_{\text{tot}}$ (say N terms).

The most straightforward way to do the rotation–translation sum in Eq. (2.5) is to simply let g run over *all* rotations (48) and translations (27 in a $3 \times 3 \times 3$-cell system.) If j' has a symmetry group S of order $O(S)$, this counts each term $O(S)$ times (as many as 48). This is unacceptably inefficient if many averages (γ's) are desired; the averaging can easily take longer than the actual molecular dynamics. Therefore we must find a way to add each distinct gj only once.

A basic (and easily verified) enumeration theorem says that all distinct gj' are obtained by taking one g from each coset $gS \subset G$ (for present purposes, time translations and inversions are not included in the group; we sum over them separately at the end). Thinking of the index set as a geometrical object, it is obvious that all translations are distinct (and easily generated by machine). So our general procedure is to enumerate the distinct rotations, and for each one generate all translations. More precisely, "distinct rotations" are defined in terms of a quotient group. Let T be the translation group. It is a normal subgroup of G, so the quotient G/T is well-defined (it is isomorphic to O_h) and there is a natural homomorphism h from G to G/T. If S' is the image of S under h, the cosets $g'S' \subset G/T$ are the "distinct rotations" of j (in crystallography, S' would be called the point group of S). Each such coset is the image of a class of cosets $gS \subset G$ which partition a set TgS (called a double coset); the double cosets partition G, and are in 1–1 correspondence with the $g'S'$. We can therefore generate a representative g of each coset $gS \subset G$, if we have a list of (arbitrarily chosen) representatives g' for all cosets $g'S' \subset G/T$: simply pick g'' so $h(g'') = g'$ (g'' may as well be a rotation about the origin; this specifies it uniquely) and generate $g \equiv tg''$ for all $t \in T$. (That we need all $t \in T$ to get all $gS \subset Tg''S$ follows because S contains no translations, hence the intersection $T \cap (g''Sg''^{-1})$ contains only the identity.)

The computer algorithm by which the averages given in Ref. 4 were obtained uses exactly the above procedure. The averages calculated so far involve eight different point groups S' (O_h, D_{4h}, C_{4v}, D_{2h}, S_2, σ_h, E). A list of coset representatives g' for each is stored (a rotation is coded by an integer from 1 to 48, which is interpreted by a subroutine which rotates indices). The inputs to the program are just $j'(\gamma)$ (coordinates of cells, etc.) and an integer label indicating its point group S'. The program computes $g''j'$ and then performs all possible translations, adding the corresponding products $\mathbf{v}_{gj'}^{\mathrm{MD}}$ computed from the molecular dynamics tape; an outer loop sums over time translations. Time-reversal symmetry is included by hand by computing averages for j' and its time-reversal image and adding.

3. EXTRACTING EQUATIONS OF MOTION

The coefficients $B_{\underline{j};\underline{k}}$ of the equation of motion (1.1) are to be computed from averages $\langle \underline{v}_{\underline{j}} \rangle$ using Eq. (1.2). In practice we must deal with cumulants $B^c_{\underline{j};\underline{k}}$ (parametrizing conditional cumulant means⁽³⁾) defined by

$$B_{\underline{j};\underline{k}} = \sum_{\underline{j}(1)\cdots\underline{k}(1)\cdots} B^c_{\underline{j}(1);\underline{k}(1)} B^c_{\underline{j}(2);\underline{k}(2)} \cdots \qquad (3.1)$$

(the sum is over all partitions $\underline{j}(1) \cup \underline{j}(2) \cup \cdots$ of $\underline{j}$, and all partitions of $\underline{k}$). Cumulant averages are defined by⁽⁶⁾

$$\langle \underline{v}_{\underline{j}} \rangle = \sum_{\underline{j}(1)\cdots} \langle \underline{v}_{\underline{j}(1)} \rangle^c \langle \underline{v}_{\underline{j}(2)} \rangle^c \cdots \tag{3.2}$$

The advantage of this is that only a finite number of the B^c are large; it also turns out that many averages which contribute strongly to Eq. (1.2) have small cumulants (i.e., are well approximated by their factorizations) and so need not be calculated in the cumulant approach.

The cumulant form of Eq. (1.2) is

$$\langle \underline{x}_{\underline{j}} \underline{h}_{\underline{k}'} \rangle^c = \sum_{\underline{k}} \sum_{\underline{j}(1)\cdots\underline{j}(1)\cdots} \sum_{\underline{k}'(1')\cdots\underline{k}(1')\cdots} B^c_{\underline{j}(1)\underline{k}(1)} B^c_{\underline{j}(2)\underline{k}(2)} \cdots \langle h_{\underline{k}'(1')} h_{\underline{k}(1')} \rangle^c \cdots \tag{3.3}$$

where the second sum is over all partitions of $\underline{k} \cup \underline{j}$ into components $\underline{k}(1) \cup \underline{j}(1), \ldots$ and the third sum is over all partitions of $\underline{k}' \cup \underline{k}$ (whose components are labeled $1', 2', \ldots$). The terms on the right-hand side are also required to be linked by k-indices (i.e., each $k \in \underline{k}$ connects one B to one $\langle \cdots \rangle$ by being a subscript of both; all factors must be indirectly connected in this fashion). This result may be proved (by induction on n, the number of indices in $\underline{j}$) by plugging Eqs. (3.1) and (3.2), and Eq. (3.3) for small n, into Eq. (1.2); unlinked terms cancel, leaving Eq. (3.3). Equation (3.3) contains an infinite number of equations, many of which are geometrically equivalent. In a practical calculation we want to use only one equation from each equivalence class; it turns out we can also avoid computing separately equivalent terms on the right-hand side. Accordingly, we classify the terms on the right side of Eq. (3.3) [each specified by the partitions $\underline{k}(1) \cdots \underline{j}(1) \cdots$ and $\underline{k}'(1') \cdots \underline{k}(1') \cdots$] into equivalence classes under rotation–translations $g \in G$, labeling the classes by integers μ. If we arbitrarily choose a representative term from each class μ whose index-set partitions $\underline{k}_\mu(1), \ldots, \underline{j}_\mu(1), \ldots, \underline{k}_\mu'(1'), \ldots, \underline{k}_\mu(1'), \ldots$ have a symmetry group S_μ, then the terms in the class are in 1–1 correspondence with cosets $gS_\mu \subset G$. The terms within a class each make the same contribution (except possibly for a sign) to Eq. (3.3), namely

$$\pm B_{\beta(1)} B_{\beta(2)} \cdots \langle \underline{v}_{\gamma(1')} \rangle \cdots \tag{3.4}$$

where we have defined $\beta(1)$ as the equivalence class of $\underline{j}_\mu(1) \cup \underline{k}_\mu(1)$ [so there is a rotation $g(1)$ so $\underline{k}_\mu(1) = g(1)\underline{j}(\beta(1))$, etc.]; similarly, $\gamma(1')$ is the class of $k'(1') \cup k(1')$ [related by the rotation $g(1')$]. (We have omitted the superscripts on B^c and $\langle v \rangle^c$; from now on everything is a cumulant.) These rotations determine the signs of the terms (3.4); the representative term has sign

$$\rho(\underline{j}(\beta(1) \cup \underline{k}(\beta(1)); g(1)) \cdots \rho(\underline{j}(\gamma(1')); g(1')) \cdots \tag{3.5}$$

and the others (associated with cosets gS_μ, $g \neq$ identity) have the additional sign factor

$$\rho(\underline{j}_\mu \cup \underline{k}_\mu'; g) \tag{3.6}$$

where $\underline{j}_\mu \equiv \underline{j}_\mu(1) \cup \underline{j}_\mu(2) \cup \cdots$, etc. These terms contribute to various different equations (3.3) (i.e., different $\underline{j} \cup \underline{k}'$); we must decide how many contribute to the equations we are interested in. The various $\underline{j} \cup \underline{k}'$ coming from a class μ are related by $g \in G$ and therefore comprise an equivalence class, say β_μ' (we omit the subscript below). Therefore we are interested in only one of them, which may be chosen to be the representative $\underline{j}(\beta') \cup \underline{k}(\beta')$ defined in Section 2, which has symmetry group $S_{\beta'}$. This may not be the same as $\underline{j}_\mu \cup \underline{k}_\mu'$; let us say it is $g_\mu \underline{j}_\mu \cup \underline{k}_\mu'$. Then the term associated with the coset $g_\mu S_\mu$ is one of the ones we want; its sign is given by Eqs. (3.5) and (3.6) with $g = g_\mu$. However, we may rotate everything by elements of $S_{\beta'}$ without changing $\underline{j} \cup \underline{k}'$ (but not by anything outside $S_{\beta'}$). Thus the cosets gS_μ corresponding to terms contributing to the equation are exactly those in the double coset $S_{\beta'} g_\mu S_\mu$. It is easy to show that these correspond to cosets $g_\mu^{-1} g S_\mu \subset (g_\mu^{-1} S_{\beta'} g_\mu)$ (note that $S_\mu \subset g_\mu^{-1} S_{\beta'} g_\mu$), so the number of such terms is exactly $O(S_{\beta'})/O(S_\mu)$. If all these terms have the same sign, this [with factors (3.5) and (3.6)] is exactly the coefficient of (3.4) in Eq. (3.3). This is in fact true; by (3.6) the sign can be changed only if some $g \in (g_\mu^{-1} S_{\beta'} g_\mu)$ changes the sign of $\underline{j}_\mu \cup \underline{k}_\mu'$, i.e., some symmetry of β' changes its sign. This of course implies [by Eq. (2.1)] that $B_{\beta'}$ vanishes by symmetry; we may simply ignore μ's that give such a β_μ'.

We may now incorporate these symmetry considerations into Eq. (3.3). For each β' (whose $B_{\beta'}$ does not vanish by symmetry) we have [using Eqs. (3.4)–(3.6)]

$$\langle \underline{v}_{\gamma(\beta')} \rangle = \sum_\mu C_\mu B_{\beta(1)} B_{\beta(2)} \cdots \langle \underline{v}_{\gamma(1')} \rangle \cdots \tag{3.7}$$

where $\gamma(\beta')$ is defined in Section 2. The sum is over all μ for which $\beta_\mu' = \beta'$ and

$$C_\mu \equiv \pm O(S_{\beta'})/O(S_\mu) \tag{3.8}$$

whose sign is Eq. (3.5) times $\rho(\underline{j}_\mu \cup \underline{k}_{\mu'}; g_\mu)$. Geometrically, C_μ is just the number of distinct rotations of μ that leave β' invariant.

Of course Eq. (3.7) still involves an infinite number of equations with an infinite number of terms. However, only a finite number affect any particular output (say one of the B's) beyond any fixed tolerance; thus the system can be solved in practice by successive truncation. Given a set of N desired B_β's, the choice of N equations to solve for them is unambiguous: clearly Eq. (3.7) should be regarded as the equation for $B_{\beta'}$. It can be seen [more easily from Eq. (3.3) than (3.7)] that the coefficient of $B_{\beta'}$ contains a product of squares $\langle h_k h_k \rangle$ which should (and in the cases calculated numerically,(4) strongly does) dominate the equation. Techniques for choosing which B's to calculate, and which terms μ to include, are discussed in Ref. 4.

The solution should clearly be done in hierarchical fashion, in order of increasing order of $\underline{j}$ (i.e., number of indices in the set $\underline{j}$.) Equations of order 1 involve only B's of order 1 (and only linearly), so these can be determined without knowing the higher order B's. In fact the j's themselves are decoupled, in the following sense: One can collect classes β into "prediction systems" according to the equivalence class γ' of $\underline{j}(\beta)$, the variable being predicted by the equation of motion. Thus for order 1 there are four γ', hence four prediction systems (energy, number, longitudinal momentum, and shear momentum transfers; only the shear transfers have been calculated numerically$^{(4)}$). Equation (3.3) does not couple these systems. Once the B's of order 1 are computed, these may be regarded as known in the equations (3.3) for the B's of order two, which are again linear equations in which the prediction systems decouple. This may be continued indefinitely; it is noteworthy that even though Eq. (3.3) is superficially nonlinear, it may be solved by purely linear methods.

One subtlety should be mentioned; Eq. (1.2) [and hence (3.3)] holds in principle for averages $\langle \underline{v}_\gamma \rangle$ calculated in *any* ensemble (whose phase space distribution depends only on cell contents and previous transfers), in particular the canonical and microcanonical ensembles. [In fact, a careful mathematical statement of the basic assumption of our theory, namely the convergence of Eq. (1.1), would probably say that Eq. (1.2) is correct for some class of "well-behaved" ensemble distribution functions.] However that does not guarantee that Eq. (1.2) determines the B's uniquely; this will only be true if the ensemble includes a complete sampling of cell histories. This is presumably true in the canonical ensemble; however, in the microcanonical ensemble, histories in which the total system contents deviate from their prescribed values are not represented at all. This leads to an ambiguity in the determination of the $B_{\underline{j};\underline{k}}$ (let us consider noncumulant B's for simplicity, although the following argument is also true for cumulants). Specifically, for each $\underline{j}$, $\underline{k}'$, and $\alpha = P, E$, or N, define a variable $\Delta_{\underline{j}\alpha\underline{k}'}$ with which to alter the B's,

$$B_{\underline{j};\underline{k}} \to B_{\underline{j};\underline{k}} + \sum \Delta_{\underline{j}\alpha\underline{k}'} \tag{3.9}$$

The sum is over the composite indices $k \in \underline{k}$ which refer to contents [so $k = (\alpha, l, 0)$], and $\underline{k}'$ is $\underline{k}$ with k omitted. It is not hard to see that in the microcanonical ensemble the new B still satisfies Eq. (1.2); this is because the coefficient of Δ involves an average with a factor $\sum_l c_{\alpha l 0}$, which vanishes identically (recall that $c_{\alpha l 0}$ is a content minus its nominal value, which we can take to be its microcanonical average). However, insisting the B's be invariant under G requires many Δ's to vanish (namely those for which some operation g preserves $\underline{j}$, α, and $\underline{k}'$ but $\rho(\underline{j}, \underline{k}; g) = -1$; this is true for all B's calculated in Ref. 4). When Δ does not vanish by symmetry, Eq. (3.9) says that there are classes of B's (generated by allowing some content variable to translate over the whole system) to which a common constant may be added. However, some of the B's in such a class should be negligible (unless the system is very small) because they involve the content of a cell far from $\underline{x}_j$. Let us choose such a distant l for each $\underline{j}$, and constrain $B_{\underline{j};\underline{k}}$ to vanish if $\underline{k}$ contains a content of cell l. This removes the ambiguity, and the equations for the remaining B's are nonsingular. This procedure defines a set of "microcanonical" B's which are different from the correct ones, but which should approach them rapidly as the system enlarges (and the true values of the B's we constrain to vanish become small).

4. INFINITE-SYSTEM LIMIT

The procedure of the previous section gives the equation-of-motion coefficients $B_{\underline{j};\underline{k}}$ for finite systems. As discussed in Section 1, these should be converted into excess-transfer parameters $\tilde{B}$. A straightforward technique for doing this is described in Ref. 4; the numerical differences are quite small. In this section we describe the extrapolation of the finite-system $\tilde{B}$'s to the infinite system.

We first compare the system-size dependence of B (or $\tilde{B}$) to that of the averages. A considerable amount is known about the system-size dependence of microcanonical averages.(7) In the dilute limit, for example, the velocity correlation function $\langle \mathbf{v}_i \mathbf{v}_j \rangle$ has a size dependence of order N^{-1} (N is the number of particles). This can be shown from the microcanonical velocity distribution function

$$P(\mathbf{v}_1 \cdots \mathbf{v}_N) = \delta\Big(\sum_i \mathbf{v}_i\Big) S(\mathbf{v}_1, \ldots, \mathbf{v}_N) \tag{4.1}$$

in which we have separated a smooth part S (a product of independent distributions of the $\mathbf{v}_i$, namely Maxwell distributions, which do not depend on system size) from the δ-function which expresses the effect of the system size through the microcanonical constraint. It follows from Eq. (4.1) that $\langle \mathbf{v}_i \mathbf{v}_j \rangle \propto -(N-1)^{-1}$; this can be understood by observing that if $\mathbf{v}_i$ takes a certain value, the other velocities are constrained to average to $-\mathbf{v}_i/(N-1)$, hence $\langle v_i v_j \rangle = -\langle v^2 \rangle/(N-1)$.

This argument can be roughly generalized to cell-variable averages. Suppose the distribution of transfers and history variables is written

$$P(x_{1/2}, h) = \prod_{\alpha} \delta\Big(\sum_l c_{\alpha l 0}\Big) S(x_{1/2}, h) \tag{4.2}$$

where the system-size effects mostly enter through the δ-functions, which express all the microcanonical constraints. We assume S is smooth and does not strongly correlate the contents (say roughly a product of Gaussians). This then has the same form as Eq. (4.1), and content averages $\langle c_{\alpha l 0} c_{\alpha l' 0} \rangle$ clearly have a term of order N^{-1}, where now N is the number of *cells*. Whether averages involving transfers also have this behavior is less clear; it is known(7) that $\langle x_{P_x,f,m}\rangle$ (the longitudinal momentum transfer, proportional to the virial pressure) does not, but this may be a special case.

The critical question now is whether the B's or $\tilde{B}$'s have the same strong N dependence. These parametrize conditional means $[\underline{x}_j]$ in ensembles of fixed history, which may be written in terms of Eq. (4.2):

$$[\underline{x}_j] = \int P(x_{1/2}, h)\underline{x}_j \, dx_{1/2} \Big/ \int P(x_{1/2}, h)\, dx_{1/2} \tag{4.3}$$

from which the δ-function (being independent of $x_{1/2}$) simply cancels, leaving the smooth S, which (we assumed) is not responsible for strong size dependence.

This argument, while not at all rigorous, strongly suggests that the B's and $\tilde{B}$'s do not have the N^{-1} size dependence of the averages, and therefore converge much faster. It is this expectation which leads us to extrapolate after, rather than before, converting the averages into B's. The expectation seems to be borne out by numerical calculations.(4)

Let us denote the $\tilde{B}$'s for a system of $M \times M \times M$ cells by $\tilde{B}^{M}_{\underline{j};\underline{k}}$. We could of course estimate the infinite-system coefficients $\tilde{B}^{\infty}$ by $\tilde{B}^{M}$ for some large M. However, there is a much more rapidly convergent technique, which involves approximately expressing $\tilde{B}^{M}$ in terms of $\tilde{B}^{\infty}$. The power-series coefficients $\tilde{B}^{M}_{\underline{j};\underline{k}}$ describe the dependence of the conditional cumulant $[\underline{x}_j]$ on the finite-system history variables: if $\underline{h}_k$ is a content of cell l, for example, $\tilde{B}^{M}_{\underline{j};\underline{k}}$ may be thought of as describing the effect on $[\underline{x}_j]$ of that particular disturbance near l that corresponds to an increase in h_k. We want to estimate $[\underline{x}_j]$ in terms of the infinite-system $\tilde{B}^{\infty}$'s. Setting $\tilde{B}^{M} = \tilde{B}^{\infty}$ would correspond to approximating the finite system by an infinite system whose history variables are h_k within a small $M \times M \times M$ region and zero outside. Clearly this ignores the periodic nature of the finite system; it would be much better to repeat the finite-system h_k in every periodic image (obtained by a translation q whose components are multiples of MW). Computing $[\underline{x}_j]$ in such a system as though it were truly infinite (i.e., using $\tilde{B}^{\infty}$) and equating the resulting power series in the finite-system h_k term by term to the correct one (involving $\tilde{B}^{M}$) gives (for $\underline{k} = k, k' \cdots$)

$$\tilde{B}^{M}_{\underline{j};\underline{k}} \approx \sum_{q,q',\cdots} \tilde{B}^{\infty}_{\underline{j};\{qk,q'k',\cdots\}} \tag{4.4}$$

where $q, q', \ldots$ range over all image translations described above. Essentially Eq. (4.4) computes the effect of a disturbance by superimposing the effect of all its images. It is straightforward to show that Eq. (4.4) has the same form for cumulant and noncumulant B's.

The above argument could also be applied to B (so that the h_k include past transfers, rather than excess transfers as implicitly assumed above), leading to a version of Eq. (4.4) without tildes. The approach we used is better if $\tilde{B}$ converges faster than B; this should be tested numerically.

Equation (4.4) is used to estimate $\tilde{B}^{\infty}$ as follows: one calculates $\tilde{B}_{\beta}{}^{M}$ for some particular M and set of β's. Writing Eq. (4.4) for each, one solves for the corresponding $\tilde{B}_{\beta}{}^{\infty}$'s. Calling these estimates $\tilde{B}_{\beta}^{\infty,M}$, we expect they converge faster than $\tilde{B}_{\beta}{}^{M}$. This is verified numerically in Ref. 4.

5. DISCUSSION

We have derived an explicit procedure for computing the infinite-fluid equation-of-motion coefficients $\tilde{B}^{\infty}$ from molecular dynamics data. From these one can compute "bare" transport coefficients, in the manner described in Ref. 3. This has been done numerically(4) for the soft-sphere liquid, obtaining some of the B's and the bare viscosity. The bare viscosity is equal to the known macroscopic viscosity(5) (within the latter's 7% statistical uncertainty) and the computation time is enormously less. This is because we extract from molecular dynamics only information about small-scale, short-lived fluctuations (the small-scale B's). The large-scale behavior, which previous methods must simulate directly at much greater expense, is treated within our formulation by an exact renormalization transformation.(3)

REFERENCES

1. P. B. Visscher, *J. Stat. Phys.* **18**:59 (1978).
2. P. B. Visscher, *J. Chem. Phys.* **68**:750 (1978).
3. P. B. Visscher, Dynamic renormalization of discrete hydrodynamics, to be published in *Physica A*.
4. Salma Begum and P. B. Visscher, *J. Stat. Phys.*, this issue, following paper.
5. W. T. Ashurst and W. G. Hoover, *Phys. Rev. A* **11**:658 (1975).
6. R. Kubo, *J. Phys. Soc. Jpn.* **17**:1100 (1962).
7. W. G. Hoover and B. J. Alder, *J. Chem. Phys.* **46**:686 (1967).