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Single-Particle Motion in the Strongly Coupled One-Component Plasma

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We present molecular dynamics computations of the time-dependent autocorrelation function of the single-particle density in a classical one-component plasma for three thermodynamic states in the range of intermediate and strong coupling. The deviations from the Gaussian approximation are calculated and the data are analyzed by the standard memory function formalism.

KEY WORDS: One-component plasma; Van Hove function; density self-correlation function; dynamical structure factor; memory function; molecular dynamics simulations.

1. INTRODUCTION

The one-component plasma (OCP) is a simplified model of very dense and completely ionized matter in which the ions are classical point charges moving in a uniform and rigid background provided by the degenerate electron gas. The model is of relevance in extreme astrophysical situations (e.g., white dwarf stars) and possibly in the laser-compressed plasmas used in fusion experiments. It has recently been the object of intense theoretical study, in particular by computer simulations. Molecular dynamics (MD) "experiments" have yielded a wealth of information on the collective modes[1] and the transport coefficients[2] of the OCP in the intermediate and strong coupling regimes. The single-particle (or "self") motion has also been studied in Ref. 1 through the computation of the velocity autocorrelation function, indicating a very strong coupling between single-particle and collective motions. However, the self part of the density autocorrelation function (or Van Hove function) had not yet been computed for nonzero wave vectors.

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It is the purpose of this paper to present MD results of this correlation function for systems of 128 and 250 ions; in particular, we have systematically computed the deviations of the correlation function from its Gaussian approximation (e.g., Ref. 3) and we have analyzed the data using the standard memory function formalism.

The paper is organized as follows: The basic definitions are recalled in Section 2. The computational technique, which is based on the expansion of the self intermediate scattering function in terms of the moments of the Van Hove function,[4] is presented in Section 3 together with the numerical results and the estimate of the statistical uncertainties, based on an exact microcanonical calculation. The memory function analysis and approximate relations between the total and self intermediate scattering functions are examined in Sections 4 and 5, and some concluding remarks are contained in the last section.

2. BASIC DEFINITIONS

We consider a system of N ions of charge Ze enclosed in a cubic volume V with periodic boundary conditions. From the mean number density $\rho = N/V$, we define our unit of length, the ion sphere radius

$$r_0 = (3/4\pi\rho)^{1/3}$$

As unit of time we choose the inverse plasma frequency ω_p^{-1}, where

$$\omega_p^2 = 4\pi\rho(Ze)^2/m$$

If the interactions are purely Coulombic, the dimensionless transport coefficients and excess thermodynamic properties depend on the single dimensionless coupling parameter

$$\Gamma = \beta(Ze)^2/r_0$$

where $\beta = 1/k_B T$.

Let $\rho_i(\mathbf{r}, t)$ be the microscopic density of the ith particle at time t:

$$\rho_i(\mathbf{r}, t) = \delta(\mathbf{r} - \mathbf{r}_i(t)) \tag{1}$$

The self part of the Van Hove function[5] is the autocorrelation function (acf) of the dynamical variable ρ_i; for a stationary, homogeneous, and isotropic fluid

$$G_s(r, t) = V\langle\rho_i(\mathbf{r}, t)\rho_i(0, 0)\rangle \tag{2}$$

The brackets denote a statistical equilibrium average. $G_s(r, t)$ is proportional to the probability density for having at time t a particle at a distance r from the origin, conditional upon its presence at the origin at $t = 0$.

The spatial Fourier transform of G_s is the self intermediate scattering function:

$$F_s(k, t) = \langle \rho_i(\mathbf{k}, t)\rho_i(-\mathbf{k}, 0)\rangle \tag{3}$$

where $\rho_i(\mathbf{k}, t) = \exp[-i\mathbf{k}\cdot\mathbf{r}_i(t)]$. We introduce the Laplace transform of the self intermediate scattering function:

$$\tilde{F}_s(k, \omega) = \int_0^\infty dt\, e^{i\omega t} F_s(k, t) \tag{4}$$

and its Fourier transform, the self dynamical structure factor:

$$S_s(k, \omega) = (1/2\pi)\int_{-\infty}^{+\infty} dt\, e^{i\omega t} F_s(k, t) \tag{5}$$

$$= (1/\pi)\tilde{F}_s'(k, \omega) \tag{6}$$

where the prime denotes the real part.

The short-time expansion of $F_s(k, t)$ reads for the OCP[1]

$$F_s(k, t) = 1 - {\omega_0}^2 t^2/2! + {\omega_0}^2\omega_{1s}^2 t^4/4! - {\omega_0}^2\omega_{2s}^4 t^6/6! + O(t^8) \tag{7}$$

where

$${\omega_0}^2 = (q^2/3\Gamma){\omega_p}^2$$

$$\omega_{1s}^2 = (q^2/\Gamma + 1/3){\omega_p}^2$$

$$\omega_{2s}^4 = (15/9)(q^4/\Gamma^2 + q^2/\Gamma) + \beta m\langle \dot{a}_{iz}^2\rangle$$

where $q = k\cdot r_0$ and $\dot{a}_{iz}$ denotes the derivative of the ith particle acceleration in the z direction.

The even moments of the Van Hove function are defined as

$$\langle r^{2p}(t)\rangle = \langle[\mathbf{r}_i(t) - \mathbf{r}_i(0)]^{2p}\rangle \tag{8}$$

From its moments the Van Hove function can be reconstructed as[4]

$$G_s(r, t) = (\alpha/\pi)^{3/2} e^{-\alpha r^2}\left[1 + \sum_{n=2}^{\infty} a_n(t) L_n^{1/2}(\alpha r^2)\right] \tag{9}$$

where $\alpha^{-1}(t) = 2\langle r^2(t)\rangle/3$ and the $L_n^{1/2}$ are Laguerre polynomials obeying the orthogonality condition

$$\int d\mathbf{r}\, e^{-\alpha r^2} L_n^{1/2}(\alpha r^2) L_{n'}^{1/2}(\alpha r^2) = \delta_{nn'}\frac{2\pi}{\alpha^{3/2}}\frac{\Gamma(n + 3/2)}{n!}$$

The coefficients $a_n(t)$ are calculated from the moments $\langle r^{2p}(t)\rangle$ by the relation

$$a_n(t) = \sum_{p=0}^{n} (-1)^p C_n^p \epsilon_p(t) \tag{10}$$

where

$$\epsilon_p(t) = [3^p/(2p+1)!!]u_p(t) - 1, \qquad \epsilon_0(t) = \epsilon_1(t) = 1 \tag{11}$$

and

$$u_p(t) = \langle r^{2p}(t)\rangle / \langle r^2(t)\rangle^p \tag{12}$$

The spatial Fourier transform of (9) yields

$$F_s(k,t) = e^{-k^2/4\alpha}\left[1 + \sum_{n=2}^{\infty}\left(\frac{k^2}{4\alpha}\right)^n \frac{a_n(t)}{n!}\right] \tag{13}$$

We introduce the well-known memory function formalism (e.g., Ref. 3, Chapter 9), which we shall use in Section 4. Let $M_s(k,t)$ [resp. $N_s(k,t)$] be the memory function associated with $F_s(k,t)$ [resp. $M_s(k,t)$]. The short-time expansion of these functions are obtained directly from (7) as

$$M_s(k,t) = \omega_0^2[1 - (\omega_{1s}^2 - \omega_0^2)t^2/2! + O(t^4)] \tag{14}$$

$$N_s(k,t) = \omega_{1s}^2 - \omega_0^2 - (\omega_{2s}^4 - \omega_{1s}^4)t^2/2! + O(t^4) \tag{15}$$

Then the normalized memory functions are defined by

$$m_s(k,t) = M_s(k,t)/\omega_0^2 \tag{16}$$

$$n_s(k,t) = N_s(k,t)/(\omega_{1s}^2 - \omega_0^2) \tag{17}$$

and we denote by $\tilde{m}_s(k,\omega)$ and $\tilde{n}_s(k,\omega)$ their Laplace transforms. We can express $\tilde{F}_s(k,\omega)$ in terms of $\tilde{m}_s(k,\omega)$ as

$$\tilde{F}_s(k,\omega) = 1/[-i\omega + \omega_0^2\tilde{m}_s(k,\omega)] \tag{18}$$

or in terms of $\tilde{n}_s(k,\omega)$ as

$$\tilde{F}_s(k,\omega) = \frac{1}{-i\omega + \omega_0^2/[-i\omega + (\omega_{1s}^2 - \omega_0^2)\tilde{n}_s(k,\omega)]} \tag{19}$$

Several approximations for $m_s(k,t)$ and $n_s(k,t)$ are considered in Section 4.

3. COMPUTATIONAL TECHNIQUE AND RESULTS

We have computed, for the three values of $\Gamma \simeq 1$, 10, and 100, the five first even moments of the Van Hove function as functions of time, from the data obtained by the MD method. From these moments, we have deduced the coefficients $a_n(t)$ with the help of (10)–(12), and then the functions $F_s(k,t)$ with the help of (13) for several values of k.

At $\Gamma \simeq 1$ (resp. $\Gamma \simeq 10$ and 100), we have simulated a periodic system of $N = 128$ (resp. $N = 250$) particles. The algorithm of Verlet[6] has been used for the integration of the classical equations of motion for the $3N$

Table I. Parameters for the MD Calculations[a]

Γ	N	N_0	Δt	T	n_c	N_c
1	128	9900	0.04	396	6	1650
9.88	250	1650	0.08	123	3	550
9.82	250	1650	0.08	123	3	550
100.	250	1800	0.3	540	1	1800

[a] N is the total number of particles. N_0 is the number of steps for each run. Δt is the time step in units of ω_p^{-1}. T is the total time of the experiment ($T = N_0 \Delta t$) in units of ω_p^{-1}. n_c is the number of time intervals between two stored configurations. N_c is the total number of stored configurations ($N_c = N_0/n_c$).

degrees of freedom, requiring the calculation of the forces between particles, and the choice of a time increment Δt. This parameter has been selected to ensure in the computations a good conservation of the total energy and to minimize the accumulation of the roundoff errors.[2] We have taken account of the long range of the Coulombic forces by the Ewald summations over the periodic replicas of the system.[7] Each experiment ran over N_0 time steps, corresponding to a total time $T = N_0 \Delta t$ (cf. Table I). The configurations were kept on magnetic tape every n_c steps (i.e., at time intervals of $\Delta t_c = n_c \Delta t$). All the parameters for each run are presented in Table I.

We obtain the moments by taking the statistical average both on the N particles and on the $N_c - n_t$ possible choices of the time origin ($t = n_t \Delta t_c$). Thus

$$\overline{r^{2p}}(t) = \frac{1}{N} \sum_{i=1}^{N} \frac{1}{N_c - n_t} \sum_{n=1}^{N_c - n_t} [\mathbf{r}_i([n + n_t] \Delta t_c) - \mathbf{r}_i(n \Delta t_c)]^{2p} \tag{20}$$

where $\overline{r^{2p}}(t)$ is the MD estimate of $\langle r^{2p}(t) \rangle$.

The first even moment $\overline{r^2}(t)$ leads to the well-known limit

$$\lim_{t \to \infty} \overline{r^2}(t) = 6Dt - 6E \tag{21}$$

where D is the self-diffusion coefficient. The values of D and E are given in Table II. The self-diffusion coefficients calculated in this way are in good agreement with those calculated from the velocity acf.[1] Because $\overline{r^2}(t)$ converges rapidly to its limit (21), we have fitted it by

$$\overline{r^2}(t) = 6E[(Dt/E) - 1 + \exp(-Dt/E)] \tag{22}$$

This fit has the disadvantage of eliminating the oscillations expected at a frequency near ω_p, since it corresponds to an exponential form for the

Table II. Values of D and E in Eq. (21), Together with M and t_c[a]

Γ	D	E	$M \times 100$	t_c
1	2.05	11.8	4.3	11
9.85	0.124	0.56	3.8	5.4
100.	6.02×10^{-3}	7.0×10^{-3}	4.94	57

[a] M is the maximum of $a_2(t)$ at $t = t_c$. D is in units of $\omega_p r_0^2$ and E in units of r_0^2.

velocity acf, which has been shown to exhibit marked oscillations[1] at $\Gamma = 10$ and 100. On the other hand, in the $t = 0$ limit, the correct behavior proportional to t^2 is correctly predicted in (22) if

$$D^2/E = 1/\beta m \tag{23}$$

This relation is effectively well verified at $\Gamma \simeq 1$, and approximately at $\Gamma = 10$ and 100.

The other moments behave for long times as

$$\overline{r^{2p}}(t) = [(2p + 1)!/p!](Dt)^p + O(t^{p-1})$$

The coefficients $a_n(t)$ are represented in Figs. 1, 2, and 3 for $\Gamma \simeq 1$, 10, and 100), respectively for $2 \leqslant n \leqslant 5$. They are affected by relatively large statistical errors, partly due to the fact that they are obtained as combinations of the "measured" ratios $\epsilon_p(t)$ [see Eqs. (10)–(12)], which lead to appreciable cancellations. We observe that for the three values of Γ, the $a_n(t)$ are always less than 10^{-2}, except for times around t_c, where $a_2(t)$ reaches its maximum M (M and t_c are given in Table II). The values of M are five to ten times smaller than in the cases of the simple liquids.[4] Note that the coefficients $a_2(t)$ vanish very slowly at long times.

We see from Figs. 1–3 that the static coefficients $a_n(t = 0)$ are nonzero. If calculated on a canonical ensemble they vanish, but on a microcanonical

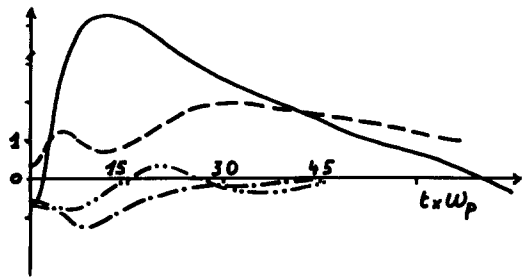

Fig. 1. Coefficients $a_n(t) \times 10^2$ at $\Gamma \simeq 1$ for $n = 2, 3, 4, 5$. (——) $a_2(t)$. (– – –) $a_3(t)$. (–·–) $a_4(t)$. (–· ·–) $a_5(t)$.

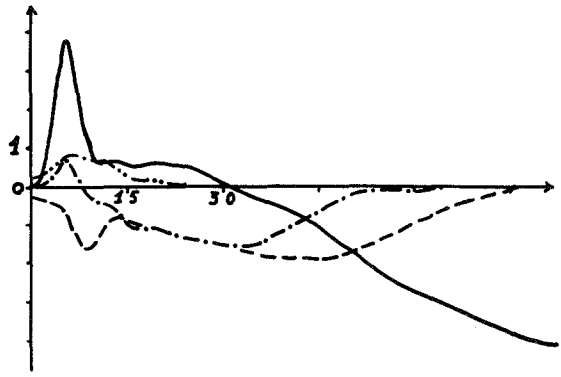

Fig. 2. Same as Fig. 1, for $\Gamma \simeq 10$.

ensemble restricted to *zero total momentum* the $a_n(0)$ are indeed nonzero. We assume that the values on the microcanonical ensemble are identical to the values calculated in a hypothetical MD experiment of infinite length. Hence we estimate the statistical uncertainties δ of the MD results by

$$\delta_n(t = 0) = |a_n^{\text{micro}}(t = 0) - a_n^{\text{MD}}(t = 0)| \tag{24}$$

We have calculated the coefficients $a_n^{\text{micro}}(t = 0)$, by introducing as an intermediary of calculation a canonical ensemble corresponding to *zero total momentum*. This allows us to separate the contributions arising from the energy fluctuations on the one hand and of the total momentum on the other hand. We have found that only the coefficient $a_2^{\text{micro}}(t = 0)$ is nonzero. The details are given in the appendix. We give in Table III the values obtained for $a_2(t = 0)$ and $\delta_n(t = 0)$.

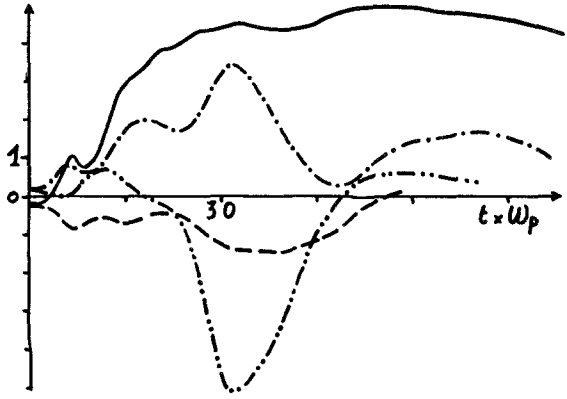

Fig. 3. Same as Fig. 1, for $\Gamma \simeq 100$.

Table III. Estimates of the Statistical Uncertainties δ_n at Time $t = 0$ for the Coefficients Given by (24)[a]

Γ	$a_2^{\text{micro}}(t=0)$	$a_2^{\text{MD}}(t=0)$	δ_2	δ_3	δ_4	δ_5
1	−4.85	−6.5	1.7	3.5	5.5	7.
10	−2.	0	2.	2.5	∼0	2.5
100	−1.3	−1.8	0.5	2.5	1.8	1.5

[a] a_2^{micro} is calculated in the Appendix [Eq. (A8)]. All values are multiplied by 100.

Consequently, for the calculation of $F_s(k, t)$, we must correct the MD data of $\overline{r^2}(t)$ and $a_n(t)$. But at short times, the corrections are negligible and at long times the errors on $F_s(k, t)$ are essentially the result of the truncation of the summation in (13). Therefore the values are correct as long as we can neglect the rest of the sum. When this is no longer justified, $F_s(k, t)$ is less than 10^{-2}, i.e., below the noise level.

We find that $F_s(k, t)$ is a decreasing function of t, with an exponential form at long times. The influence of the collective modes is drowned in the purely diffusive character of the single-particle motion.

Since the coefficients $a_n(t)$ are very small, the Gaussian approximation for $F_s(k, t)$ is excellent: we denote by $F_s^G(k, t)$ the corresponding function. We denote by $F_s^{GF}(k, t)$ the same approximation in which we replace $\overline{r^2}(t)$ by the fit (22). The analysis of these approximations is made on their Laplace transform $\tilde{F}_s(k, \omega)$. We have collected all the results in Section 4 (see Tables IV–VI and Figs. 4–9).

4. ANALYSIS OF $\tilde{F}_s(k, \omega)$ FROM THE MEMORY FUNCTION FORMALISM

We approximate by an exponential or Gaussian form the normalized memory functions introduced in (16) and (17). We measure the accuracy of these approximations compared to "the exact $\tilde{F}_s^{\text{MD}}(k, \omega)$, obtained from the MD data, by calculating the root mean square deviation δ with respect to the value $\tilde{F}_s^{\text{MD}}(k, \omega)$:

$$\delta = \left[\frac{1}{\tilde{F}_s^{\text{MD}}(k, 0)\omega_{\text{MAX}}} \int_0^{\omega_{\text{MAX}}} [\tilde{F}_s^{\text{MD}}(k, \omega) - \tilde{F}_s(k, \omega)]^2 \, d\omega \right]^{1/2} \tag{25}$$

where ω_{MAX} is taken equal to $2\omega_p$. The results of these calculations are presented in Tables IV, V, and VI) for $\Gamma \simeq 1$, 10, and 100, respectively.

Table IV. Deviation of the Approximate Value of $\tilde{F}_s(k, \omega)$ from the Exact Value, for Various Values of k, for $\Gamma \simeq 1$[a]

q	G	GF	ME	MG	MGF	NE	NG	NGF
0.2	3.1	3.2	5.5	110	4.5	1.5	31	3.0
0.4	1.7	2.0	7.0	29	1.6	0.4	27	2.8
0.6	0.9	1.1	6.3	9.3	0.3	1.5	17	1.4
1.	0.7	1.	10.	2.1	0.9	5.6	11	0.7
1.4	0.4	0.8	11.	2.	1.5	7.6	28	0.9
1.9	0.2	0.7	12.	3.5	1.8	8.9	29	1.2

[a] The deviation is measured by 100δ, where δ is given by (25). The headings of the columns refer to the various approximations defined in Section 4.

Table V. Deviation of the Approximate Value of $\tilde{F}_s(k, \omega)$ from the Exact Value, for Various Values of k, for $\Gamma \simeq 10$[a]

q	G	GF	ME	MG	MGF	NE	NG	NGF
0.3	2.	1.7	1.8	180	5.4	4.1	8	3.2
0.6	3.6	3.9	3.8	72	9.1	7.8	13.	6.3
0.9	2.0	3.3	2.1	38	7.4	7.1	14.	4.7
2.3	0.7	4.9	5.3	1.4	1.1	6.3	5.8	1.4
4.	0.6	8.1	10.3	3.2	1.4	7.9	1.8	1.2
6.	0.2	8.4	11.	4.2	1.7	9.3	5.5	1.4

[a] See footnote to Table IV.

Table VI. Deviation of the Approximate Value of $\tilde{F}_s(k, \omega)$ from the Exact Value, for Various Values of k, for $\Gamma \simeq 100$[a]

q	G	GF	ME	MG	MGF	NE	NG	NGF
0.6	0.09	0.06	0.8	144	0.06	1.1	0.8	0.4
0.9	0.8	0.3	1.1	82	0.3	2.2	1.6	1.0
1.2	2.5	2.0	2.2	60	1.6	3.4	3.4	2.1
1.9	6.6	6.0	5.8	42	3.8	3.4	7.2	3.1
3.	6.	5.5	8.2	23	5.3	4.4	6.0	4.2
6.	1.5	1.7	7.8	7.9	1.7	7.0	2.1	1.9

[a] See footnote to Table IV.

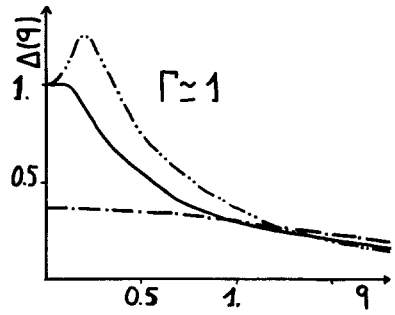

Fig. 4. Plot of $\Delta(q)$ for $\Gamma \simeq 1$, using the various approximations defined in the text ($q = k \cdot r_0$). (——) MD data; the ME, MGF, NE, NGF, G, and GF results are practically indistinguishable from these MD data. (–· ·–) MG approximation). (–··–) NG approximation.

Two other interesting quantities are the values of $\tilde{F}_s(k, 0)$ and the half-width at half-height $\omega_{1/2}$ of $\tilde{F}_s(k, \omega)$. We consider the following dimensionless quantities[8]:

$$\Sigma(k) = k^2 D \tilde{F}_s(k, 0) \tag{26}$$

$$\Delta(k) = \omega_{1/2}/k^2 D \tag{27}$$

which go to one in the hydrodynamic limit $k = 0$. The quantities $\Sigma(k)$ and $\Delta(k)$ are represented in Figs. 4–9.

The short-time behavior of the following approximations being well known,[3] we pay particular attention to their variations as a function of ω.

(*i*) *Exponential form for* $m_s(k, t)$: We characterize this function by a single relaxation time $\tau_m(k)$ depending only on k,

$$m_s(k, t) = \exp[-t/\tau_m(k)] \tag{28}$$

$\tau_m(k)$ is chosen so as to yield the "exact" limit $\omega = 0$ for $\tilde{F}_s^{\mathrm{ME}}(k, \omega)$. Thus we have

$$\tau_m(k) = \beta m D/\Sigma^{\mathrm{MD}}(k) = \omega_0^2/\tilde{F}_s^{\mathrm{MD}}(k, 0)$$

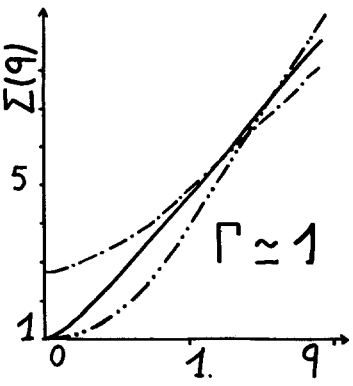

Fig. 5. For $\Gamma \simeq 1$, plot of $\Sigma(q)$, using the various approximations defined in the text ($q = k \cdot r_0$). The key is the same as in Fig. 4.

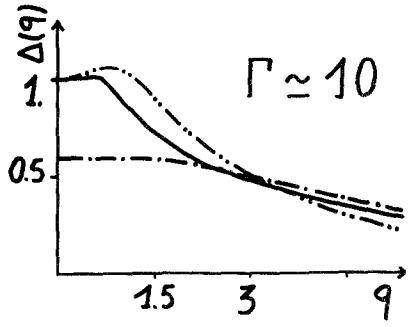

Fig. 6. Same as Fig. 4, for $\Gamma \simeq 10$.

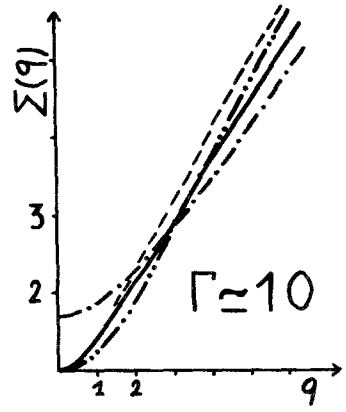

Fig. 7. Same as Fig. 5, for $\Gamma \simeq 10$; (---) GF approximation.

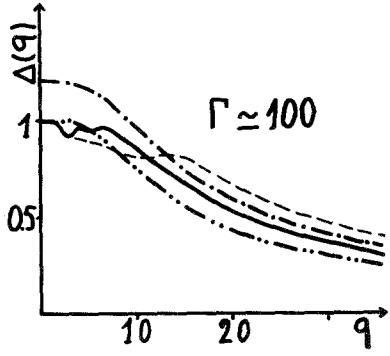

Fig. 8. Same as Fig. 4, for $\Gamma \simeq 100$; (---) NE approximation.

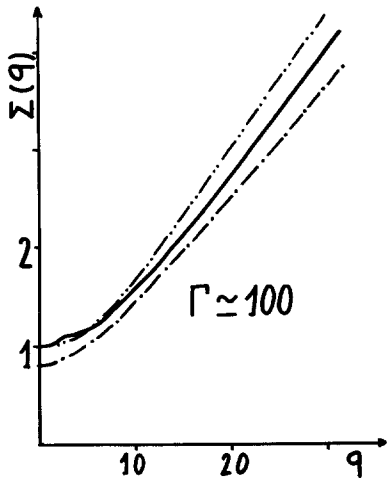

Fig. 9. Same as Fig. 5, for $\Gamma \simeq 100$.

By definition $\Sigma^{ME}(k)$ is identical with $\Sigma^{MD}(k)$. The variations of $\Delta^{ME}(k)$ reproduce qualitatively those of $\Delta^{MD}(k)$.

(ii) Exponential form for $n_s(k, t)$: In a way similar to (28), we write

$$n_s(k, t) = \exp[-t/\tau_n(k)] \tag{29}$$

and $\tau_n(k)$ is again selected to give the "exact" limit $\omega = 0$ for $\tilde{F}_s^{NE}(k, \omega)$, so that

$$\tau_n(k) = \Sigma_{(k)}^{MD}/[\beta m D(\omega_{1s}^2 - \omega_0^2)] = \tilde{F}_s^{MD}(k, 0)/(\omega_0^2\omega_{1s}^2 - \omega_0^4)$$

The values obtained for $\Sigma^{NE}(k)$ are still equal to $\Sigma^{MD}(k)$. The variations of $\Delta^{NE}(k)$ are very close to the MD results.

(iii) Gaussian form for $m_s(k, t)$: We now assume a Gaussian form for $m_s(k, t)$:

$$m_s(k, t) = \exp[-\tfrac{1}{2}B_m(k)t^2] \tag{30}$$

We express $B_m(k)$ in two distinct ways. Either from $\Sigma^{MD}(k)$, as in the two preceding cases [we note $\tilde{F}_s^{MGF}(k, \omega)$], or from the exact behavior at short times, by Eq. (14) (we write $\tilde{F}_s^{MG}(k, \omega)$]:

$$B_m^{MGF}(k) = \tfrac{1}{2}\pi[\Sigma^{MD}(k)/\beta m D]^2 = \tfrac{1}{2}\pi[\omega_0^2\tilde{F}_s^{MD}(k, \omega)]^2 \tag{31}$$

$$B_m^{MG}(k) = \omega_{1s}^2 - \omega_0^2 \tag{32}$$

In the second case, we do not obtain the correct hydrodynamic limit for $\Sigma^{MG}(k)$. But values obtained for $\Sigma^{MGF}(k)$ and $\Delta^{MGF}(k)$ are excellent.

(*iv*) *Gaussian form for* $n_s(k, t)$: We use the following form:

$$n_s(k, t) = \exp[-\tfrac{1}{2}B_n(k)t^2] \tag{33}$$

As in the preceding case, we have two ways to express $B_n(k)$. The "exact" limit $\omega = 0$ for $\tilde{F}^{\mathrm{NGF}}(k, \omega)$ defines $B_n^{\mathrm{NGF}}(k)$ as

$$B_n^{\mathrm{NGF}}(k) = \frac{\pi}{2}\left[\frac{(\omega_{1s}^2 - \omega_0^2)\beta mD}{\Sigma^{\mathrm{MD}}(k)}\right]^2 = \frac{\pi}{2}\left[\frac{\omega_{1s}^2 - \omega_0^2}{\omega_0^2 \tilde{F}_s^{\mathrm{MD}}(k, 0)}\right]^2 \tag{34}$$

By definition, $\Sigma^{\mathrm{NGF}}(k)$ is still equal to $\Sigma^{\mathrm{MD}}(k)$. The variations of $\Delta^{\mathrm{NGF}}(k)$ are almost identical to those of $\Delta^{\mathrm{MD}}(k)$.

The other choice for B_n is determined by the exact behavior at short times, from (17):

$$B_n^{\mathrm{NG}}(k) = (\omega_{2s}^4 - \omega_{1s}^4)/(\omega_{1s}^2 - \omega_0^2)$$

By taking this Gaussian form for $n_s(k, t)$. Vieillefosse and Hansen[9] expressed the diffusion coefficient for the OCP as

$$\beta mD = 1/[\Omega_0^2 \tilde{n}_s(0, 0)] = (1/\Omega_0^2)[2B_n^{\mathrm{NG}}(0)/\pi]^{1/2}$$

where $\Omega_0^2 = \omega_p^2/3$. Inversely, from the diffusion coefficient, we calculate B_n^{NG}. For nonzero k, we have[4]

$$B_n^{\mathrm{NG}}(k) = \frac{B_n^{\mathrm{NG}}(0) + 3\omega_0^2(3 + 2\omega_0^2/\Omega_0^2)}{1 + 2\omega_0^2/\Omega_0^2} \tag{35}$$

The corresponding values of $\Sigma^{\mathrm{NG}}(k)$ are too small at small k and too large at large k.

In conclusion, the better approximations are those that reproduce the "exact" $\omega = 0$ limit for $\tilde{F}_s(k, \omega)$ (ME, NE, MGF, NGF). For these approximations the variations of $\Delta(k)$ are quantitatively near their MD values. But some differences arise for $\omega \sim \frac{1}{2}\omega_{1/2}$ and $\omega \sim 2\omega_{1/2}$. The former are essentially accounted for by δ (see Tables IV–VI), while the latter yield different behaviors for $\tilde{F}(k, \omega)$ as shown in the next section.

5. RELATIONS BETWEEN $\tilde{F}_s(k, \omega)$ AND $\tilde{F}(k, \omega)$

Different theories allow us to relate $\tilde{F}_s(k, \omega)$ and $\tilde{F}(k, \omega)$, the total intermediate scattering function. The memory function formalism allows us to derive relations between $\tilde{F}_s(k, \omega)$ and $\tilde{F}(k, \omega)$ (Ref. 3, Chapter 9) by introducing some approximations between their memory functions. The Vineyard approximation[10] amounts to equalizing the memory functions $M_s(k, t)$ and $M(k, t)$, and leads to the relation

$$\tilde{F}(k, \omega) = S(k)\tilde{F}_s(k, \omega)$$

where $S(k)$ is the static structure factor. This relation is incapable of yielding the plasmon mode at the small values of k.

In the second approximation, by imposing the correct small-time behavior between the memory functions

$$M_s(k, t) = S(k)M(k, t)$$

we obtain the relation obtained by Kerr[11]:

$$\tilde{F}(k, \omega) = \frac{S(k)\tilde{F}_s(k, \omega)}{1 - [S(k) - 1/S(k)[i\omega\tilde{F}_s(k, \omega) - 1]} \tag{36}$$

The precision of the computations does not allow the use of the MD results of $\tilde{F}_s^{MD}(k, \omega)$ to test this relation, because for $\omega \sim \omega_{1/2}$, $\tilde{F}_s^{MD}(k, \omega)$ is very small. Hence we have replaced $\tilde{F}_s(k, \omega)$ in (36) by the various approximations introduced in Section 4 and we have compared the results with the "exact" data for $S(k, \omega) = (1/\pi)\tilde{F}'(k, \omega)$ obtained by Hansen *et al.*[1] The best agreement is achieved with the NE approximation [exponential form for

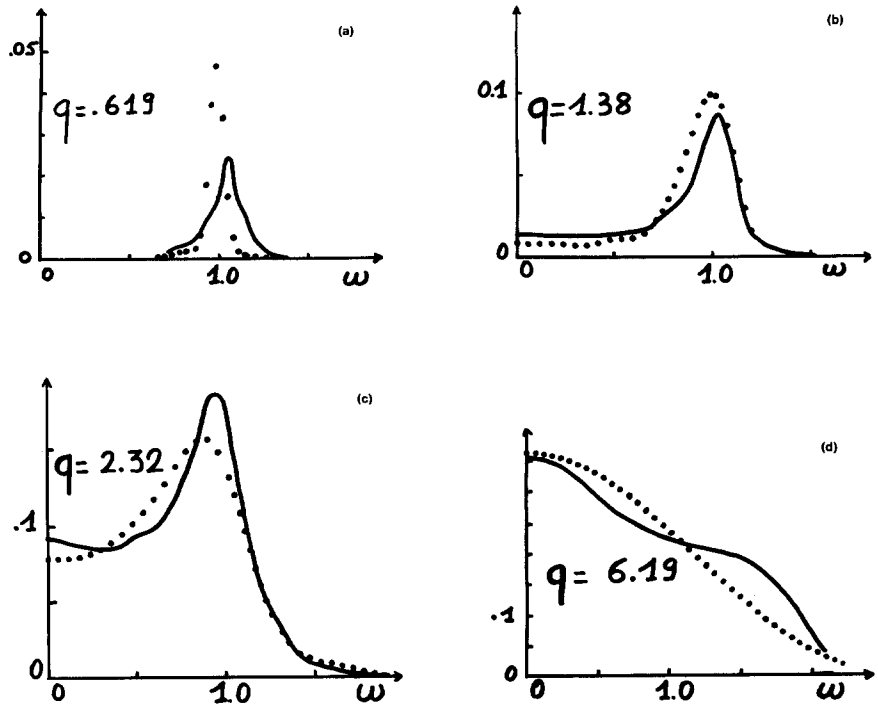

Fig. 10. Comparison at $\Gamma \simeq 10$ between the exact dynamical structure factor (dotted line) and the one obtained from Eq. (36) with the NE approximation for $F_s(k, \omega)$ [Eq. (29)] (solid line), for four values of $q = k \cdot r_0$.

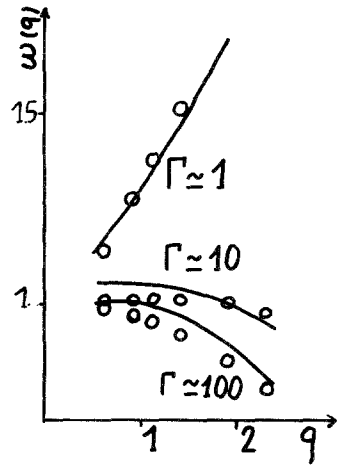

Fig. 11. Dispersion curves at $\Gamma \simeq 1, 10, 100$ obtained from the approximate NE structure factor [Eqs. (36) and (29)] (solid line) and exact results (dots) for $\Gamma \simeq 1, 10, 110$. Here ω is in units of ω_p and q in units of r_0^{-1}.

$n_s(k, t)$], especially for small k. The MGF and NGF approximations give equally good results for $q = kr_0 > 1$.

In Fig. 10, we represent, for $\Gamma \simeq 10$, $S^{\mathrm{NE}}(k, \omega)$ for four values of k. In Fig. 11, we give the dispersion curves for $\Gamma \simeq 1$, 10, 100 obtained from the same NE approximation. The agreement with the "exact" MD data of Ref. 1 is good.

6. CONCLUSION

We have shown that in the strong coupling regime, the deviations from the Gaussian behavior of the function $F_s(k, t)$ are weaker than in the case of simple liquids. In particular, we do not observe a plasmon peak in $\tilde{F}_s(k, \omega)$, the influence of the collective motions being less perceptible than in the spectrum of the velocity acf $Z(t)$.[1] This is not very surprising since it is well known that in the $k \to 0$ limit

$$Z(t) = -\lim_{k \to 0} \frac{1}{k^2} \frac{d^2}{dt^2} F_s(k, t)$$

This means that upon integrating an oscillating $Z(t)$ twice with respect to time, the oscillations are essentially smoothed out and their amplitude in $F_s(k, t)$ is reduced to below the noise level. An estimate of the statistical uncertainties has been obtained by an exact calculation in the microcanonical ensemble.

The analysis of the results based on the memory function formalism shows that a better accuracy is achieved by imposing the "exact" $\omega = 0$ limit for the function $\tilde{F}_s(k, \omega)$, which contains a global information, rather than by imposing the exact short-time behavior.

We have also examined the link between the total and self dynamical structure factors. Good agreement is achieved once more by using the self functions that satisfy the exact $\omega = 0$ limit. The best results are obtained at small k by the NE approximation [Eqs. (36) and (29)].

APPENDIX

We calculate, on a microcanonical ensemble characterized by a total energy E_0 and zero total momentum, the mean value $\bar{A}(E_0)$ of a function $A(\mathbf{r}_N, \mathbf{p}_N)$ of the positions and velocities of the N particles of the system. We denote by $\omega(E)$ the volume of phase space of energy E and zero total momentum:

$$\omega(E) = \int d\mathbf{r}_N \, d\mathbf{p}_N \, \delta(H(\mathbf{r}_N, \mathbf{p}_N) - E) \, \delta\left(\sum_{i=1}^{N} \mathbf{p}_i\right) \tag{A1}$$

The mean value of A on a canonical ensemble at the temperature T, with the restriction of zero total momentum, is defined as

$$\langle A \rangle_{T,\mathbf{p}=0} = \frac{\int e^{-\beta E} \omega(E) \bar{A}(E) \, dE}{\int e^{-\beta E} \omega(E) \, dE} \tag{A2}$$

We choose to define the temperature T of the canonical ensemble from E_0 by the implicit relation

$$\langle H \rangle_{T,\mathbf{p}=0} = E_0 \tag{A3}$$

We now assume that the function $\bar{A}(E)$ varies slowly and expand $\bar{A}(E)$ to second order around E_0; the first-order term vanishes, due to (A3), and we find

$$\langle A \rangle_{T,\mathbf{p}=0} = \underset{(\alpha)}{\bar{A}(E_0)} + \underset{(\beta)}{\frac{1}{2!} \frac{d^2 \bar{A}(E_0)}{dE^2}} \underset{(\gamma)}{\langle (E - E_0)^2 \rangle_{T,\mathbf{p}=0}} + \underset{(\delta)}{\cdots} \tag{A4}$$

(α) and (β) are of the same order of magnitude, while $(\gamma)/(\alpha)$ is of order $1/N$ and $(\delta)/(\alpha)$ of order $1/N^2$. Hence in (γ) we may replace all the quantities by their canonical ensemble values [neglecting terms of order $(\alpha)/N^2$], then (A4) can be rewritten as

$$\bar{A}(E_0) = \langle A \rangle_{T,\mathbf{p}=0} - \frac{1}{2!} k_B T^2 C_v(T) \frac{d^2 \langle A \rangle_T}{d \langle H \rangle_T^2} \tag{A5}$$

where $C_v(T)$ is the total specific heat at constant volume. For the calculation of $\langle A \rangle_{T,\mathbf{p}=0}$, we suppose in addition that the interaction potential does not

depend on the velocities, and the function $A(\mathbf{r}_N, \mathbf{p}_N)$ has the form $A_1(\mathbf{r}_N)A_2(\mathbf{p}_N)$. Hence the first term on the rhs of (A5) separates into

$$\langle A\rangle_{T,\mathbf{p}=0} = \langle A_1\rangle_T \langle A_2\rangle_{T,\mathbf{p}=0} \tag{A6}$$

Consequently E_0 can be expressed from the excess canonical internal energy $\langle V\rangle_T$ as

$$E_0 = (N-1)\tfrac{3}{2}k_B T + \langle V\rangle_T$$

For the OCP, $\langle V\rangle_T$ is very well fitted by the following expression due to De Witt[12]:

$$(1/Nk_B T)\langle V\rangle_T = a\Gamma + b\Gamma^{1/4} + c$$

with $a = -0.896434$, $b = 0.861852$, and $c = -0.55513$.

As an application of (A5) and (A6), we calculate the coefficient $a_2(t)$ to second order, and the other coefficients to zeroth order. We first express the moments $\overline{r^{2p}}(t)$ in the short-time limit as functions of microcanonical static expressions:

$$\overline{r^2}(t) = \overline{\mathbf{V}_i^2}(0)t^2 + \left[\frac{1}{4}\frac{\overline{\mathbf{F}_i^2(0)}}{m^2} + \frac{1}{3}\overline{\mathbf{V}_i(0)\cdot\frac{d^2\mathbf{V}_i(0)}{dt^2}}\right]t^4 + O(t^6)$$

A straightforward calculation yields

$$\overline{\mathbf{V}_i^{2p}} = \langle \mathbf{V}_i^{2p}\rangle_T\left\{1 - \frac{p}{N} - \frac{k_B}{C_v(T)}\left[\frac{p(p-1)}{2} - \frac{p}{2}\frac{d\log C_v(T)}{d\log T}\right]\right\} \tag{A7}$$

From this we derive the coefficients at $t = 0$

$$a_2^{\text{micro}}(0) = -k_B/C_v(T), \qquad a_p^{\text{micro}}(0) = 0 \quad \text{if } p \geqslant 3 \tag{A8}$$

The coefficient $a_2(t)$ estimated on a canonical ensemble is proportional to t^4 at short times.[13] For the OCP we give its expression to second order in time and first order in $1/N$:

$$a_2^{\text{micro}}(t) = -\frac{1}{N}\left[\frac{Nk_B}{C_v(T)} + \frac{1}{18}\frac{(\omega_p t)^2}{\xi''} + O\left(t^4, \frac{1}{N}\right)\right] \tag{A9}$$

where

$$\xi'' = -\frac{1}{N}\frac{d\beta\langle H\rangle_T}{d\log\Gamma} = \frac{9}{2} + \frac{3\beta\langle H\rangle_T}{N} - 9\beta\left.\frac{\partial p}{\partial \rho}\right|_T \tag{A10}$$

The values of ξ'' estimated from the MD data have the same sign as ξ'' in (A10). But the order of magnitude is difficult to estimate because the first calculated values of $a_2(t)$ are for $t = 0.12$ at $\Gamma \simeq 1$ and 0.3 at $\Gamma \simeq 100$ and higher order terms in the expansion (A9) are probably nonnegligible. Nevertheless, this order of magnitude appears to be correctly reproduced by the MD data.

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