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Microscopic theory of self-diffusion in a classical one-component plasma

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We apply the fully renormalized kinetic-theory formalism of Mazenko to the study of self-diffusion in a dense one-component classical plasma. The memory function associated with the phase-space correlation function for self-diffusion is expressed in terms of an effective two-body problem which allows us to make approximations at a microscopic level in a straightforward manner. We use simple physical arguments to obtain the "effective interaction approximation" for the memory function in which a pair of particles interact via a dynamically screened effective potential. This approximation for the memory function is a nontrivial and natural generalization of the Balescu-Guernsey-Lenard form that includes the exact statics of the system, treats screening at large and small distances consistently, and is valid for all wave numbers and frequencies. The inclusion of the hydrodynamic modes in the memory function leads to an oscillatory long-time tail in the velocity autocorrelation function. We calculate the self-diffusion constant as a function of density using the effective interaction form of the memory function and calculate the memory function associated with the velocity autocorrelation function using an interpolation procedure which incorporates the hydrodynamic modes. Qualitative agreement with recent molecular-dynamics computations of the self-diffusion constant and the memory function is found.

I. INTRODUCTION

We present in this paper a new approach to the microscopic calculation of the time-dependent equilibrium-averaged correlation functions of a dense one-component classical plasma. The "one-component classical plasma" (OCP) is an electrically neutral system of charged classical particles of one species embedded in a uniform background of opposite charge. The background does not contribute to the dynamics. The dense OCP provides an excellent model for understanding many features of stellar[1] interiors and laser-compressed plasmas[2] and has interesting properties and complications associated with it due to the long-range nature of the Coulomb potential.

The properties of an OCP can be characterized by the dimensionless parameter[3] Γ defined by

$$\Gamma = e^2/k_B Ta\,, \tag{1.1}$$

where $a=(3/4\pi n)^{1/3}$ and n is the number density. The "weakly coupled" OCP and dense OCP are characterized by $\Gamma \ll 1$ and $\Gamma > 1$, respectively.

The time correlation functions of an OCP for $\Gamma \ll 1$ can be obtained from the linearized Balescu-Guernsey-Lenard[4] (BGL) kinetic equation. However the range of validity of this equation and of the modifications which remove the short-range divergence[4] of the BGL collision integral are restricted to small wave vector k and small frequency ω as well as to $\Gamma \ll 1$. One goal of the present work is to obtain a linearized kinetic equation which is valid for all k and ω and which is applicable to a dense plasma. The "dense" or strongly coupled OCP of interest here lacks a small parameter in the usual perturbation theoretic sense.

We restrict ourselves in this paper to an investigation of the self-motion of the particles and consider the velocity autocorrelation function

$$V_D(t) = \tfrac{1}{3}\langle \vec{V}_1(t)\cdot\vec{V}_1(0)\rangle \tag{1.2}$$

giving the correlation of a tagged particle's (particle 1, say) velocity at time t with its initial velocity. In Eq. (1.2) $\langle\ \rangle$ denotes an average over an equilibrium ensemble. We shall also consider the self-diffusion constant D which can be determined from the time integral[5] of $V_D(t)$:

$$D=\int_0^\infty dt\ V_D(t)\,. \tag{1.3}$$

It is useful to analyze the time dependence of $V_D(t)$ by writing its rate of decay in terms of the "memory function" $M(t)$:

$$\frac{dV_D(t)}{dt} = -\int_0^t dt'\,M(t')\,V_D(t-t')\,. \tag{1.4}$$

Hansen, McDonald, and Pollack[6,7] have reported extensive molecular-dynamics (MD) computations of $V_D(t)$, and D for the OCP over a wide range of Γ. The most striking feature of their results is a coupling of the single-particle motions to the collective density fluctuations. This coupling is manifested at $\Gamma \gtrsim 10$ and at long times in the appearance of oscillations in $V_D(t)$ and $M(t)$ at a frequency near the plasma frequency $\omega_p=(4\pi ne^2/m)^{1/2}$. We present a simple microscopic theory which yields the oscillatory behavior of $V_D(t)$ and $M(t)$ for long times. We also calculate $M(t)$ and D as a function of Γ, and obtain qualitative agreement with the MD computations of $M(t)$ and D. A brief report[8] of some of the results of this work has been published.

The microscopic theory is based on the fully renormalized kinetic-theory[9-11] (FRKT) formalism developed by Mazenko, whose notation we adopt. Because of the complications associated with the hard-sphere systems considered in Refs. 9–11, the present application of the FRKT formalism to the OCP might serve as a relatively simple introduction to the formalism. The OCP is also a useful model for the comparison of the results of various approximations made within the FRKT approach with "experiment" (MD computations), since the parameter Γ which characterizes the OCP ranges from less than unity to a maximum value[12] of approximately 155 at which the OCP crystallizes. In contrast to the OCP the analogous dimensionless parameter for a hard-sphere system has a maximum value of approximately 0.5 corresponding to close packing. (The dimensionless parameter can be taken to be equal to $\pi n\sigma^3/6$, where σ is the hard-sphere diameter.)

Although the correlation function of interest is $V_D(t)$, it is convenient to consider a generalization of V_D for which the momentum and spatial coordiantes of a tagged particle are treated on a equal basis. The starting point of the formalism is the exact kinetic equation satisfied by this two-point phase-space correlation function. This equation defines a collision integral or memory function which represents the effect of the other $N-1$ particles in the system on the tagged particle. The main features of the FRKT formalism are the expression of the memory function as an effective two-body problem, the systematic inclusion of the exact static correlation functions, and the analysis of the correlation functions in terms of their connected and disconnected parts. The formal reduction of the N-body problem to an effective two-body problem allows us to introduce approximations for the memory function in a natural way and as part of a systematic and well-defined approximation procedure in which the terms that have been neglected are known in principle. The present analysis allows us to separate the intrinsically static effects from the dynamical effects; the inclusion of the exact static correlations is especially important in an OCP since the long-range static correlations dominate the dynamics in an essential way. In contrast to the present method another approach[13] to dense plasmas is the decomposition, consistent with the sum rules and general symmetry conditions, of higher-order correlation functions in terms of lower-order correlation functions. This approach leads to nonlinear equations which are to be solved for both the static and dynamic properties of the system. The disadvantage of this approach is that the limitations of the approximations are difficult to determine and the nonlinear equations are difficult to solve.

In Sec. II A we introduce the phase-space correlation functions of interest for self-diffusion and relate the velocity autocorrelation function to the two-point phase-space correlation function C_s. We introduce the memory function ϕ_s in Sec. II B and obtain an exact expression for ϕ_s in terms of a four-point correlation function which represents the dynamical correlation of two particles in the medium. This form for ϕ_s leads us in Sec. III to make the "disconnected approximation" in which only the disconnected part of the four-point correlation function is retained. This leads to an approximation for the memory function that can be interpreted as the physical process in which a pair of particles interact, propagate independently through the medium, and recollide with each other. In Sec. IV A we make contact with previous work by neglecting the presence of the medium on the pair of "intermediate" particles. This approximation leads to a form for ϕ_s which is not useful for Coulomb systems. We include effects of the medium in Sec. IV B and assume that the intermediate particles move in an average field due to all the other particles. This assumption yields the "effective-interaction-approximation" from for ϕ_s in which a pair of particles interact via a dynamically screened effective potential. This form for ϕ_s is a nontrivial and natural generalization of the BGL form for ϕ_s to finite values of k and ω and to finite values of Γ. In Sec. IV C we obtain the long-time behavior of $V_D(t)$ by including the hydrodynamic modes associated with the collisional effects of the medium and find that $V_D(t)$ is proportional to $t^{-3/2}\sin\omega_p t$. The qualitative behavior of this long-time tail is in agreement with the MD calculation[6] of $V_D(t)$.

We discuss the properties of the effective interaction in Sec. V A. In Sec. V B we relate the self-diffusion constant D to the time integral of the memory function $M(t)$ and calculate D as a function of Γ using the effective-interaction approximation for $M(t)$. [The relation of $M(t)$ to ϕ_s is given in Appendix A.] The Monte Carlo results[14] for the two-particle static structure function are used as input. The calculated values of D exhibit a rapid variation in Γ consistent with the MD values of D. In Sec. V C we approximate the intermediate correlation functions with forms which interpolate between their mean-field behavior and their long-time hydrodynamic behavior. The resultant form for $M(t)$ is evaluated numerically and found to be in qualitative agreement with the MD results for $M(t)$ except for $t\approx 0$, where the disconnected approximation breaks down because of its incorrect treatment of close collisions.

We discuss the qualitative features of our analysis in Sec. VI and indicate areas of future work.

II. FORMALISM

A. Definitions

We present a review of the FRKT[9-11] in order to emphasize its physical features and mathematical structure. Consider a system of N particles of mass m in a volume Ω with the density $n=N/\Omega$ a fixed constant. Since we are interested in the correlation of the motion of a tagged particle (particle 1) with itself and the other particles in the system, we define the phase-space densities

$$f_s(1)=N^{1/2}\delta(1-q_i)\,, \tag{2.1}$$

and

$$f(1)=\sum_{i=1}^{N}\delta(1-q_i)\,, \tag{2.2}$$

where $q_i\equiv(R_i,P_i)$ are the phase-space coordinates of particle i and $1\equiv(r_1,p_1)$. The velocity autocorrelation function can be obtained from the time-dependent phase-space self-correlation function

$$C_s(12,t-t')=\langle\delta f_s(1,t)\delta f_s(2,t')\rangle\,, \tag{2.3}$$

where $f_s(1,t)=e^{iLt}f_s(1)$, L is the Liouville operator, and δf_s is the deviation of f_s from its equilibrium value. We work with the spatial Fourier transform

$$C_s(k,p_1p_2,t)=\int\frac{d^3r_1\,d^3r_2}{\Omega}\,e^{i\vec{k}\cdot(\vec{r}_1-\vec{r}_2)}\,C_s(12,t)\,, \tag{2.4}$$

the temporal Fourier transform

$$C_s(12,\omega)=\int_{-\infty}^{+\infty}dt\;e^{+i\omega t}C_s(12,t)\,, \tag{2.5}$$

and the temporal Laplace transform

$$\begin{aligned}C_s(12,z)\equiv C_s(12)&=-i\int_0^{+\infty}dt\;e^{izt}C_s(12,t)\\&=\int_{-\infty}^{+\infty}\frac{d\omega'}{2\pi}\,\frac{C_s(12,\omega')}{z-\omega'}\,.\end{aligned} \tag{2.6}$$

The relation $C_s(12,\omega)=-2\,\mathrm{Im}C_s(12,z=\omega+i0^+)$ follows from (2.6). In the following ω will denote a real frequency, z a complex frequency with $\mathrm{Im}z>0$, and the z dependence of $C_s(12,z)$ will often be omitted. The dynamical van Hove self-correlation function $S_s(k,\omega)$, the Fourier transform in space and time of the correlation in position of a tagged particle with its initial position, is simply related to C_s by

$$n\,S_s(k,\omega)=\int d^3p_1\,d^3p_2\,C_s(k,p_1p_2,\omega)\,. \tag{2.7}$$

Similarly $V_D(t)$, the velocity autocorrelation function defined in (1.2), is given as

$$V_D(t)=\frac{1}{3m^2n}\int d^3p_1\,d^3p_2\,\vec{p}_1\cdot\vec{p}_2\,C_s(k=0,p_1p_2,t)\,. \tag{2.8}$$

We shall also require the time-dependent phase-space density correlation function

$$C(12,t)=\langle\delta f(1,t)\delta f(2,0)\rangle\,, \tag{2.9}$$

which is the phase-space generalization of the dynamic structure function $S(k,\omega)$,

$$nS(k,\omega)=\int d^3p_1\,d^3p_2\,C(k,p_1p_2,\omega)\,, \tag{2.10}$$

with the spatial and temporal Fourier transform of (2.9) defined as in (2.4) and (2.5). In (2.9), $\delta f(1)$ is the deviation of $f(1)$ from its equilibrium value,

$$\begin{aligned}\langle f(1)\rangle&=nf_0(p_1)\\&=n(\beta/2\pi m)^{3/2}\exp(-\beta p_1^2/2m)\,.\end{aligned} \tag{2.11}$$

Other dynamical functions of interest are the three- and four-point correlation functions for self-diffusion,

$$C_s(1,23,t)=\langle\delta f_s(1,t)\delta(f_s(2,0)f(3,0))\rangle\,, \tag{2.12}$$

$$C_s(12,34,t)=\langle\delta(f_s(1,t)f(2,t))\delta(f_s(3,0)f(4,0))\rangle\,. \tag{2.13}$$

The exact static properties of the system enter into the formalism through the initial value of the time-dependent correlation functions. Momentum-dependent static correlation functions will be denoted by a tilde, e.g., $\tilde{C}_s(12)\equiv C_s(12,t=0)$. In the thermodynamic limit we have

$$\tilde{C}_s(12)=n\delta(12)f_0(p_1)\,, \tag{2.14}$$

and

$$\tilde{C}(12)=n\delta(12)f_0(p_1)+n^2f_0(p_1)f_0(p_2)h(r_1-r_2)\,, \tag{2.15}$$

where

$$h(r)=g(r)-1\,, \tag{2.16}$$

and the static pair distribution function $g(r)$ is defined by

$$n^2g(r_1-r_2)=\left\langle\sum_{i\neq j}^{N}\delta(r_1-R_i)\delta(r_2-R_j)\right\rangle\,. \tag{2.17}$$

A comparison of (2.14) and (2.15) shows that the static correlations for the density fluctuations are more complicated than for self-diffusion. This relative complexity implies that $S(k,\omega)$ exhibits a much richer structure than its counterpart $S_s(k,\omega)$. The momentum integral of $\tilde{C}(12)$ is related to the static structure function $S(r)$ by

$$nS(r_1-r_2)=\int d^3p_1\,d^3p_2\,\tilde{C}(12)$$
$$=n\delta(r_1-r_2)+n^2h(r_1-r_2)\,. \tag{2.18}$$

It is also useful to introduce the direct correlation function C_D defined in terms of the Fourier transform of $h(r)$ as

$$C_D(k)=h(k)[1+nh(k)]^{-1}\,. \tag{2.19}$$

It will be important in the following to analyze the correlation functions in terms of their disconnected and connected parts since the former dominates the latter contribution at large particle separation. Because $C_s(12,34)$ possesses many disconnected pieces, we are led to define a new four-point correlation function

$$G_s(12,34)=C_s(12,34)-C_s(12,\bar{2})C_s^{-1}(\bar{2}\bar{3})C_s(\bar{3},34)\,, \tag{2.20}$$

where $C_s(12,34)$ and $C_s(12,3)$ are the Laplace transforms of (2.13) and (2.12), respectively, and the inverse C_s^{-1} is defined by

$$C_s^{-1}(1\bar{1})C_s(\bar{1}2)=\delta(12)\,. \tag{2.21}$$

[Repeated indices such as $\bar{2}$ in (2.20) imply an integration[15] over that variable.] The disconnected part of G_s is given by

$$G_{sD}(12,34,t)=C_s(13,t)C(24,t)\,. \tag{2.22}$$

It is possible to define an inverse of G_s such that

$$G_s^{-1}(12,\bar{1}\bar{2})G_s(\bar{1}\bar{2},34)=\delta(13)\delta(24)\,. \tag{2.23}$$

Note that in (2.22) and (2.23) there is no "exchange" term[16] $(1\leftrightarrow 2, 3\leftrightarrow 4)$ for G_s in contrast to its presence in the corresponding quantity for density fluctuations. The difference between the two quantities is that in G_s the indices 1 and 3 are associated with the tagged particle, and the indices 2 and 4 are associated with all N particles in the system.

B. Collisional effects

We treat collisional effects on the time evolution of $C_s(12)$ by introducing the function $\varphi_s(12)$, which gives the modification of the free-particle motion of a tagged particle due to the other $N-1$ particles. The memory function φ_s is defined by the generalized kinetic equation[17]

$$\left(z-\frac{\vec{\mathrm{k}}\cdot\vec{\mathrm{p}}}{m}\right)C_s(k,pp',z)-\int d^3\bar{p}\,\varphi_s(k,p\bar{p},z)C_s(k,\bar{p}p',z)$$
$$=\tilde{C}_s(k,pp') \tag{2.24}$$

Free-particle streaming is given by the first term on the left-hand side of (2.24). It is seen that φ_s has the physical interpretation of a nonlocal, non-Markoffian source function modifying the free-particle motion; such an interpretation leads to the description of φ_s as the memory function for the self-diffusion correlation function. In field-theory language φ_s renormalizes the single-particle motion. The memory function $\varphi(12)$ for the density fluctuations can be defined in the same manner by the generalized kinetic equation for $C(12)$:

$$\left(z-\frac{\vec{\mathrm{k}}\cdot\vec{\mathrm{p}}}{m}\right)C(k,pp',z)-\int d^3\bar{p}\,\varphi(k,p\bar{p},z)C(k,\bar{p}p',z)$$
$$=\tilde{C}(k,pp')\,. \tag{2.25}$$

In general, φ_s and φ can be separated into a z-independent (static) part and z-dependent (collisional) part, e.g.,

$$\varphi(k,pp',z)=\varphi^{(s)}(k,pp')+\varphi^{(c)}(k,pp',z)\,. \tag{2.26}$$

The static part can be evaluated without approximation.[11] As an illustration of the more complicated static correlations in (2.25) compared to (2.24),

$$\varphi^{(s)}(k,pp')=-(\vec{\mathrm{k}}\cdot\vec{\mathrm{p}}/m)C_D(k)nf_0(p)\,, \tag{2.27}$$

and the static part of φ_s vanishes identically. Note that $\varphi^{(s)}$ is independent of p'.

The evaluation of φ_s is the major problem in developing a microscopic theory. Since φ_s represents the effects of the medium on a tagged particle, it is not possible in general to obtain φ_s exactly. We can make progress by formulating φ_s in terms of an effective two-body problem. Since such a formulation has been given in Ref. 11 for the more complicated case of density fluctuations, we only outline the method here. If (2.24), which defines φ_s, is compared to the usual equation of motion[18] which relates $C_s(12)$ to $C_s(1,23)$, we can obtain φ_s in terms of $C_s(1,23)$. We then use the equation of motion satisfied by $C_s(1,23)$ to obtain a formal expression for φ_s in terms of $G_s(12,34)$. The result is

$$\varphi_s(12)nf_0(p_1)=-\int d\bar{1}\,d\bar{2}L_I(1\bar{1})L_I(2\bar{2})G_s(1\bar{1};2\bar{2})\,, \tag{2.28}$$

where $L_I(12)=i\nabla_{r_1}V(r_1-r_2)\cdot(\nabla_{p_1}-\nabla_{p_2})$ is the interaction part of the two-particle Liouville operator. The form (2.28) is symmetrical and is in the form of an effective two-body problem in which $G_s(1\bar{1},2\bar{2})$ represents the dynamical evolution of two particles, labeled by phase-space coordinates 1 and $\bar{1}$, where particle 1 is tagged and $\bar{1}$ is not. We wish to replace the "bare" interactions $L_I(1\bar{1})$ between these particles in (2.28) by an effective interaction which is determined by the static correlations. We "separate out" the static correlation in G_s and write

$$G_s(12,34)=\tilde{G}_s(12,\bar{1}\bar{2})\overline{G}_s(\bar{1}\bar{2},\bar{3}\bar{4})\tilde{G}_s(\bar{3}\bar{4}:34)\,, \tag{2.29}$$

which defines the new four-point function $\overline{G}_s$. If we combine (2.28) and (2.29) we find the desired form

$$\varphi_s(12)nf_0(p_2)=-\,V_s(1,\bar{1}\bar{2})\overline{G}_s(\bar{1}\bar{2},\bar{3}\bar{4})V_s(2,\bar{3}\bar{4})\,, \tag{2.30}$$

where the "end-point" vertices are

$$V_s(1,23)=\int d\bar{2}\,L_I(1\bar{2})\tilde{G}_s(1\bar{2},23)\,. \tag{2.31}$$

V_s is completely determined by the static correlations of the system and, using the techniques developed in the Appendix of Ref. 11, can be shown to be given by

$$V_s(1,23)=-\,n^2f_0(p_2)f_0(p_3)g(r_2-r_3)\tilde{L}_I(23)\delta(12)\,, \tag{2.32}$$

where

$$\tilde{L}_I(23)=i\beta^{-1}[\nabla_{r_2}\ln g(r_2-r_3)]\cdot(\nabla_{p_2}-\nabla_{p_3})\,. \tag{2.33}$$

$\tilde{L}_I$ can be interpreted as a renormalized two-particle Liouville interaction operator. Note that V_s is missing an "exchange" term which is present in the end-point vertex for density correlations.[11]

III. DISCONNECTED APPROXIMATION

A. Form of φ_s

We discuss a simple approximation for $\overline{G}_s$ which emphasizes the collective properties characteristic of a plasma and which leads to a realistic first approximation for φ_s. Recall that $\overline{G}_s$ represents the dynamical correlation of two particles, one of which is tagged, and consider the process in which two particles are initially well separated but interacting, propagate independently of each other, and then reinteract. That is, in the intermediate state, we ignore the mutual interactions between the two particles and emphasize their interaction with the remaining $N-2$ particles in the medium. These qualitative considerations imply that we approximate $\overline{G}_s$ by its disconnected part $\overline{G}_{sD}$ [see (2.22) and (2.29)]:

$$\begin{aligned}\overline{G}_s(11',22',t)&\approx\overline{G}_{sD}(11',22'\ t)\\&=\tilde{C}_s^{-1}(1\bar{1})\tilde{C}_s^{-1}(2\bar{2})\tilde{C}^{-1}(1'\bar{4})\tilde{C}^{-1}(2'\bar{3})\\&\quad\times C_s(\bar{1}\bar{2},t)C(\bar{4}\bar{3},t)\,.\end{aligned} \tag{3.1}$$

(Recall that integration over repeated barred indices is implied.) For consistency we have had to approximate the static correlation function $\tilde{G}_s(12;34)$ by its disconnected part to obtain the disconnected part of $\overline{G}_s$ from G_s. C_s and C in (3.1) are the exact correlation functions given by (2.24) and (2.25), respectively, and include the interaction of the particles with the medium.

It is clear from the form of (3.1) that $\overline{G}_{sD}$ represents the independent motion of two particles. We refer to (3.1) as the disconnected approximation—it is the basic approximation of this paper. This approximation, which has been described qualitatively here, can be made part of a systematic approximation procedure[11] in which the additional contributions to $\overline{G}_s$ and hence φ_s can be calculated. The major process that has been neglected in the disconnected approximation is dynamically uncorrelated binary collisions of the type treated by the Boltzmann equation, in which the two particles are initially localized and propagate mostly under their mutual interaction. It might be expected that this process would be important for short times and for large Γ. We will delay discussion of the contribution of close collisions to future work.

We gain additional insight into the physical interpretation of (3.1) by obtaining the explicit form of φ_s implied by (3.1). If we substitute (3.1) in (2.30) and use the fact that $\tilde{C}_s^{-1}(12)=\delta(1-2)$ $[nf_0(p_1)]^{-1}$, we have

$$\varphi_s(12)nf_0(p_2)=-\,\overline{V}_s(1,\bar{1}\bar{2})C_s(\bar{1}\bar{3})C(\bar{2}\bar{4})\overline{V}_s(2,\bar{3}\bar{4})\,, \tag{3.2}$$

where

$$\overline{V}_s(1,1'2')=V_s(1,1'\bar{2})\tilde{C}^{-1}(\bar{2}2')/nf_0(p_{1'})\,. \tag{3.3}$$

The analysis of φ_s is conveniently performed in terms of the spatial Fourier transform

$$\begin{aligned}\overline{V}_s(1,1'2')=\int\frac{d^3k}{(2\pi)^3}\frac{d^3q}{(2\pi)^3}&\exp[+i\vec{\mathrm{k}}\cdot(\vec{\mathrm{r}}_1-\vec{\mathrm{r}}_{1'})]\\&\times\exp[i\vec{\mathrm{q}}\cdot(\vec{\mathrm{r}}_{1'}-\vec{\mathrm{r}}_{2'})]V_s(kq,p_1p_{1'}p_{2'})\,.\end{aligned} \tag{3.4}$$

From (2.32), (3.3), and the form

$$\tilde{C}^{-1}(k,p_1p_2)=\delta(p_1-p_2)/nf_0(p_2)-C_D(k)\,, \tag{3.5}$$

we find the simple result

$$\overline{V}_s(kq,p_1p_2p_3)=-\,\beta^{-1}C_D(q)\vec{\mathrm{q}}\cdot\nabla_{p_1}\delta(p_1-p_2)\,. \tag{3.6}$$

Note that $\overline{V}_s(kq,p_1p_2p_3)$ is independent of k and p_3

and depends on p_2 in a trivial manner. We take the spatial Fourier transform of (3.2), substitute in (3.6), perform the momentum integrations implied in (3.2), and obtain the form

$$\varphi_s(k,p_1p_2,t)nf_0(p_2) = -n^2\int\frac{d^3q}{(2\pi)^3}[\beta^{-1}C_D(q)]^2\vec{q}\cdot\nabla_{p_1}\vec{q}\cdot\nabla_{p_2} \times C_s(k-q,p_1p_2,t)S(q,t)\,, \qquad (3.7)$$

where $S(q,t)$ is the temporal Fourier transform of (2.10). The form (3.7) for φ_s is a direct consequence of (3.1). It is seen from (3.7) that the disconnected approximation implies that the tagged particle's motion is coupled to the density fluctuations in a simple manner.

B. Effective-interaction approximation

The main quantity of interest is not the phase-space memory function φ_s but the velocity autocorrelation function as given by (2.8). From (2.24) it is seen that owing to the coupling between the momentum indices it is necessary to solve a nonlinear integral equation for C_s and then to integrate the solution over the momenta. We can solve (2.24) using the method of kinetic modeling.[19] This method consists of replacing (2.24) by a series of approximations where, presumably, the series eventually converges to the exact solution. It is shown in Appendix A that in the simplest approximation (equivalent to the one-Sonine-polynomial approximation) the solution to (2.24) yields the following approximate relation for the memory function of the velocity autocorrelation function in terms of the phase-space memory function:

$$M_{1p}(t)=\frac{1}{3n(mV_0)^2} \times\int d^3p_1\,d^3p_2\,\vec{p}_1\cdot\vec{p}_2\varphi_s(k=0,p_1p_2,t)f_0(p_2)\,. \qquad (3.8)$$

In the above $mV_0^2=k_BT$ and the subscript $1p$ indicates the one-Sonine-polynomial approximation. If we insert (3.7) for φ_s in (3.8) we obtain M_{1p} in terms of $S_s(q,t)$ and $S(q,t)$. In order to preserve the exact static behavior of $S(q,t)$ we write

$$S(q,t)=S(q)G(q,t)\,. \qquad (3.9)$$

We note that $S_s(q,t=0)=G(q,t=0)=1$. If we substitute (3.7) in (3.8) and use (3.9) we obtain

$$M_{1p}(t)=\frac{n}{3(mV_0)^2}\int\frac{d^3q}{(2\pi)^3}q^2[\beta^{-1}C_D(q)]^2 \times S(q)S_s(q,t)G(q,t)\,. \qquad (3.10)$$

We emphasize that (3.10) follows directly from two approximations, i.e., (3.1) and an approximate solution of the kinetic equation (2.24).

The $t=0$ limit of (3.10) can be written using (2.16) and (2.19) in the form

$$M_{1p}(t=0)=-\frac{1}{3m}\int\frac{d^3q}{(2\pi)^3}q^2[-\beta^{-1}C_D(q)][S(q)-1]\,. \qquad (3.11)$$

The form (3.11) is convenient for showing the breakdown of the disconnected approximation at very short times, since the exact result[20] for $M(t=0)$ is

$$M(t=0)=-\frac{1}{3m}\int\frac{d^3q}{(2\pi)^3}q^2V(q)[S(q)-1]=\frac{\omega_p^2}{3}\,. \qquad (3.12)$$

The quantity $V(q)$ is the Fourier transform of the interparticle potential. Because of the uniform background which makes the OCP electrically neutral, the integral in (3.12) is identically equal to $\omega_p^2/3$. Note that in the limit $\Gamma\ll 1$ the quantity $-\beta^{-1}C_D(q)\rightarrow V(q)$ and (3.11) agrees with (3.12). The discrepancy between (3.11) and (3.12) is not due to the neglect of close collisions (large q) but to their incorrect treatment; the integral over q in (3.11) is convergent for both large and small q because of the consistent inclusion of the exact statics.

IV. INTERMEDIATE CORRELATION FUNCTIONS

The disconnected approximation was shown in Sec. III to lead to a form of the memory function φ_s in terms of the intermediate propagators C_s and S. In principle, the form of the intermediate propagators should be obtained from a self-consistent solution of (2.24) and (3.7) for C_s and a similiar set of equations for C. In lieu of such a sophisticated approach, we show that the use of relatively simple forms for the intermediate correlation functions leads to reasonable results for φ_s and $M(t)$, gives us some insight into the level of complexity of microscopic calculations, and indicates areas where further work is necessary.

A. Free-particle approximation

The simplest approximation for the intermediate correlation functions is to assume that the "intermediate" particles are free. This "free-particle" approximation leads to a result for φ_s which is not useful for Coulomb systems. We consider it here in order to make contact with previous work and to illustrate the simplicity of the method.

The free-particle forms for S_s and G can be found by solving (2.24) and (2.25) with $\varphi_s=\varphi^{(s)}=\varphi^{(c)}=0$. The solutions are

$$C_s(q,p_1p_2,z)=nf_0(p_1)\delta(p_1-p_2)(z-\vec{q}\cdot\vec{p}_1/m)^{-1} \qquad (4.1)$$

and

$$G(q,z)=Q(q,z)\,, \tag{4.2}$$

where

$$Q(q,z)=\int d^3p f_0(p)(z-\vec{q}\cdot\vec{p}/m)^{-1}\,. \tag{4.3}$$

We take the Laplace transform of (3.7), substitute in (4.1)–(4.3) and obtain

$$\varphi_s^{\rm FP}(k,pp',z)f_0(p')=\frac{n}{(mV_0)^2}\int\frac{d^3q}{(2\pi)^3}\int d^3\bar{p}\,[\beta^{-1}C_D(q)]^2S(q)\,\vec{q}\cdot\nabla_p\vec{q}\cdot\nabla_{p'}\left(\frac{f_0(p)f_0(\bar{p})\delta(p-p')}{z-(\vec{k}-\vec{q})\cdot\vec{p}/m-\vec{q}\cdot\vec{\bar{p}}/m}\right). \tag{4.4}$$

The static structure function $S(q)$ in (4.4) is a measure of the initial correlation of the two particles; their effective interaction is $-\beta^{-1}C_D(q)$. For a weakly coupled system the interparticle potential is nonsingular and much smaller in magnitude than β^{-1}. In this limit $-\beta^{-1}C_D(q)\to V(q)$, $S(q)\to 1$, and (4.4) reduces to the results of Ref. 17 for the case of self-diffusion; in the additional limits of small k and z, $\varphi_s^{\rm FP}$ reduces to the linearized Fokker-Planck form.[4]

The form of $M_{1p}(t)$ corresponding to (4.4) is easily seen to be

$$M_{1p}^{\rm FP}(t)=\frac{n}{3(mV_0)^2}\int\frac{d^3q}{(2\pi)^3}q^2[\beta^{-1}C_D(q)]^2 \times S(q)\exp(-q^2V_0^2t^2)\,. \tag{4.5}$$

The long-time behavior of $M_{1p}^{\rm FP}(t)$ for an OCP is of interest and is determined by the behavior of the integrand of (4.5) for $q\ll(V_0t)^{-1}$. The small-q behavior of the exact $S(q)$ and $C_D(q)$ are given by

$$S(q)\xrightarrow[q\to 0]{}q^2/4\pi n\beta e^2 \tag{4.6}$$

and

$$-\beta^{-1}C_D(q)\xrightarrow[q\to 0]{}4\pi e^2/q^2\,. \tag{4.7}$$

If we use (4.6) and (4.7) in (4.5), we find that the long-time ($t\gg\omega_p^{-1}$) behavior of $M_{1p}^{\rm FP}(t)$ for an OCP is given by

$$M_{1p}^{\rm FP}(t)\to(e^2/6)(m\beta^3/\pi)^{1/2}t^{-3}\,. \tag{4.8}$$

As will be discussed in Sec. V. $M^{\rm FP}$ yields results for the self-diffusion constant D of an OCP which are in disagreement by two orders of magnitude with the MD computations of D. Thus we conclude that although $M^{\rm FP}$ contains the exact statics, the dynamical approximations that lead to (4.4) and (4.5) are not adequate for a system with long-range correlations.

B. Effective interaction approximation (EIA)

The simplest realistic approximation for the intermediate untagged particle is that it moves in an average field due to all the other particles. This assumption is equivalent to setting $\varphi^{(c)}=0$ and retaining only the static term $\varphi^{(s)}$ in (2.24). The resultant equation for $C(k,pp',z)$ can be interpreted as a linearized Vlasov equation[4] and solved exactly[21] to give $G(q,z)$ as

$$G(q,z)=Q(q,z)/\epsilon(q,z)\,, \tag{4.9}$$

where $\epsilon(q,z)$ is the dielectric function generalized to retain the exact statics and is given by

$$\epsilon(q,z)=\{1+[S(q)-1]zQ(q,z)\}/S(q)\,. \tag{4.10}$$

The corresponding approximation for the intermediate correlation function of the tagged particle is the free particle form (4.1), since the static part of φ_s vanishes identically. We substitute (4.9) and (4.1) in the Laplace transform of (3.7) and use the fact that for a stable system $S(q,z)$ is analytic in the upper half z plane to find

$$\varphi_s^{\rm EIA}(k,pp',z)f_0(p')=\frac{n}{(mV_0)^2}\int\frac{d^3q}{(2\pi)^3}S(q)[\beta^{-1}C_D(q)]^2\vec{q}\cdot\nabla_p\vec{q}\cdot\nabla_{p'}\frac{Q(q,z-(\vec{k}-\vec{q})\cdot\vec{p}/m)f_0(p)\delta(p-p')}{\epsilon(q,z-(\vec{k}-\vec{q})\cdot\vec{p}/m)}. \tag{4.11}$$

We can use the identity $Q/\epsilon=[Q+(S-1)z\,|Q^2|1/S|\epsilon|^2$ to write (4.10) in the physically transparent form

$$\varphi_s^{\rm EIA}(k,pp',z)f_0(p')=\frac{n}{(mV_0)^2}\int\frac{d^3q}{(2\pi)^3}\,\vec{q}\cdot\nabla_p\vec{q}\cdot\nabla_{p'}\left|\frac{\beta^{-1}C_D(q)}{\epsilon(q,z-(\vec{k}-\vec{q})\cdot\vec{p}/m)}\right|^2 \times\{Q(q,z-(\vec{k}-\vec{q})\cdot\vec{p}/m)+z[S(q)-1]\,|Q(q,z-(\vec{k}-\vec{q})\cdot\vec{p}/m)|^2\}f_0(p)\delta(p-p')\,. \tag{4.12}$$

The quantity $-\beta^{-1}C_D/|\epsilon|$ can be interpreted as the effective interaction and is discussed in Sec. V A. The form (4.11) will be referred to as the "effective-interaction" approximation[22] for φ_s.

It is easy to see that in the limit $\Gamma<1$, a weakly coupled OCP for which $S(q)$ and $C_D(q)$ reduce to their Debye-Hückel forms,[4] and for small k and z, $\varphi_s^{\rm EIA}$ reduces to the well-known linearized

Balescu-Guernsey-Lenard (BGL) form.[4] Hence, (4.11) is a natural generalization of the BGL form for φ_s which retains the exact statics of the system and is valid for all k and z.

Because of the presence of the exact static correlation functions, φ_s^{EIA} is free of a short-range divergence[23] in contrast to the BGL form of φ_s. However, as discussed in Sec. III, the integrand in (4.11) is not correct for large q because of the use of the disconnected approximation.

We postpone a discussion of the time-dependence of $M^{\mathrm{EIA}}(t)$ to Sec. V and state here the result (5.12) that for long times $M^{\mathrm{EIA}}(t) \sim t^{-3}\cos\omega_p t$. The $\cos\omega_p t$ factor arises from the zero of ϵ associated with the long-wavelength collective behavior of the plasma oscillations. It is also shown that M^{EIA} yields numerical results for the self-diffusion constant which are in qualitative agreement with the molecular-dynamics computations for a wide range of Γ.

C. Long-time limit

The effective-interaction approximation for $M(t)$ gives reasonable values for D but does not give the long-time behavior of $M(t)$ correctly since the mean-field approximations for S_s and G neglect the hydrodynamical modes that dominate the long-time behavior of the exact S_s and G, and consequently the long-time behavior of $M(t)$. The hydrodynamic behavior (small q, large t) of $S_s(q,t)$ and $G(q,t)$ is known exactly[24] and is given by

$$S_s(q,t) = \exp(-q^2 D t) \tag{4.13}$$

$$G(q,t) = (1 - C_V/C_P)\exp(-q^2 D_T t) + \frac{C_V}{C_P}\cos\omega_p t \exp(-q^2\Gamma_{nn} t/2)\,, \tag{4.14}$$

where

$$\Gamma_{nn} = \frac{4/3\eta + \zeta}{mn} + D_T\left(\frac{C_P}{C_V} - 1\right)\,. \tag{4.15}$$

In the above C_P and C_V are the specific heats, η is the shear viscosity, ζ is the bulk viscosity, and D is the self-diffusion coefficient. D_T is the thermal diffusion coefficient and is related to the thermal conductivity λ by $D_T = \lambda/mn\,C_P$. Note the presence of the $\cos\omega_p t$ factor in (4.13). MD results[14] show that for $\Gamma = 1$ the ratio $\gamma = C_P/C_V$ differs from unity by ~3%; for $\Gamma \geq 20$ the difference is less than 1% and decreases with increasing Γ. Thus, in the following we set $C_P = C_V$ and combine (3.10), (4.13), and (4.14) to obtain

$$M_{1p}^{H}(t) = \frac{n}{3(mV_0)^2}\int\frac{d^3q}{(2\pi)^3}\,S(q)[\beta^{-1}C_D(q)]^2 q^2 \times \exp[-q^2(D+\Gamma_{nn}/2)t]\cos\omega_p t\,. \tag{4.16}$$

The inclusion of the hydrodynamic behavior of the intermediate propagators in (4.16) is equivalent to the ideas used in mode-mode coupling theories. The form (4.16) represents the coupling of the single-particle motion to the long-lived, long-wave length density fluctuations in the system.

We can evaluate the long-time behavior of (4.16) without approximation for an OCP since the long-time behavior is dominated by the small-q behavior of the integrand. In the limit of small q the exact $S(q)$ and $C_D(q)$ are given by (4.7) and (4.6), respectively.[14] The result is

$$M_{1p}(t) \to \frac{\sqrt{2}\,e^2}{3\pi m(D+\Gamma_{nn}/2)^{3/2}}\,t^{-3/2}\cos\omega_p t\,. \tag{4.17}$$

The result (4.17) is the exact long-time limit of (3.10). We see from (4.17) that for long times the coupling of the motion of single particles to the long-wavelength density fluctuations leads to a long-time tail which oscillates about zero with a frequency ω_p is qualitative agreement with the MD computation of the "exact" $M(t)$. The corresponding long-time behavior of $V_D(t)$ is shown in Appendix B to be

$$V_D(t) \to \frac{-3k_B T(D+\Gamma_{nn}/2)^{3/2}}{2^{3/2}\omega_p e^2}\,t^{-3/2}\sin\omega_p t\,. \tag{4.18}$$

Note that D and Γ_{nn} appearing in (4.17) and (4.18) are the exact transport coefficients.

The behavior of $M(t)$ for intermediate times is considered in Sec. V C.

V. NUMERICAL EVALUATION

We now calculate the memory function $M(t)$ and the self-diffusion constant D of a one-component classical plasma within the disconnected approximation using an iterative approach. The effective-interaction approximation M^{EIA} is chosen as the zeroth-order approximation to $M(t)$ and used to obtain the zeroth order or "bare" D. This choice for the zeroth-order approximation is motivated by the fact that M^{EIA} contains the essential physics of the OCP, i.e., screening and plasma oscillations, retains the exact statics of the system, and reduces to well-known results in the limit of small Γ. We next consider the contributions to $M(t)$ arising from the inclusion of the hydrodynamic modes.

A. Effective interaction

In Sec. IV B it was pointed out that the quantity

$$\Phi(q,\omega) = -\,\beta^{-1}C_D(q)/\left|\,\epsilon(q, z = \omega + i0^+)\right| \tag{5.1}$$

can be interpreted as an effective frequency- and wave-vector-dependent interparticle potential. The static ($\omega = 0$) behavior of Φ can be obtained from (4.10) and is given by $\Phi(q) \equiv \Phi(q,0) = -\beta^{-1}C_D(q)S(q)$. $\Phi(q)$ is screened at large distances as can be seen from its exact small-g behavior determined by (4.6) and (4.7), that is, $\Phi(0) = (n\beta)^{-1}$. The spatial Fourier transform of $\Phi(q)$ can be written as $\Phi(r)/k_\beta T = 1 - g(r)$. The Debye-Hückel approximation for $\Phi(r)$ is $\Phi_{\rm DH}(r) = (e^2/r)e^{-r/D}$, where $D^2 = (4\pi n e^2 \beta)^{-1}$. A comparison of $\Phi(r)$ using the Monte Carlo values of $g(r)$ as input with $\Phi_{\rm DH}(r)$ is shown in Fig. 1 for $\Gamma = 20$. It is seen that $\Phi(r)$ is bounded for all r and approaches a constant as $r \to 0$ in contrast to the behavior of $\Phi_{\rm DH}(r)$.

To analyze the behavior of $\Phi(q,\omega)$ for $\omega \neq 0$ we first define the plasma dispersion function[25]

$$Z(\nu) = \sqrt{\pi} \int_{-\infty}^{+\infty} dx \frac{e^{-x^2}}{x - \nu - i0^+} \equiv -2F(\nu) + iB(\nu)\,, \tag{5.2}$$

where

$$F(\nu) = e^{-\nu^2} \int_0^\nu dx\, e^{x^2} \tag{5.3}$$

and

$$B(\nu) = \sqrt{\pi}\, e^{-\nu^2}\,. \tag{5.4}$$

$F(\nu)$ is frequently referred to as Dawson's integral.[25] The dielectric function can be related to Z by using (4.3), (4.10), and (5.2) to obtain

$$|\epsilon(k, \nu + i0^+)| = D(k,\nu)/S(k) \tag{5.5}$$

where

$$D^2(k,\nu) = \{1 + 2[S(k) - 1]\,\nu F(\nu)\}^2 + \{[S(k) - 1]\,B(\nu)\}^2\,. \tag{5.6}$$

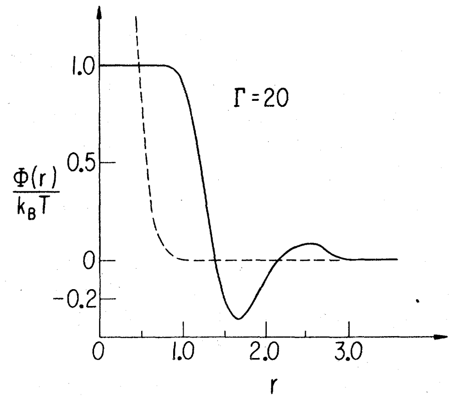

FIG. 1. The (dimensionless) static effective interaction as a function of the interparticle distance r (in units of a) for $\Gamma = 20$. The solid line represents $\Phi(r)/k_B T = 1 - g(r)$, with the Monte Carlo values of $g(r)$ as input. The dashed line represents the Debye-Hückel approximation $\Phi_{\rm DH}(r)/k_B T = (\Gamma/r)\exp[-(3\Gamma)^{1/2} r]$.

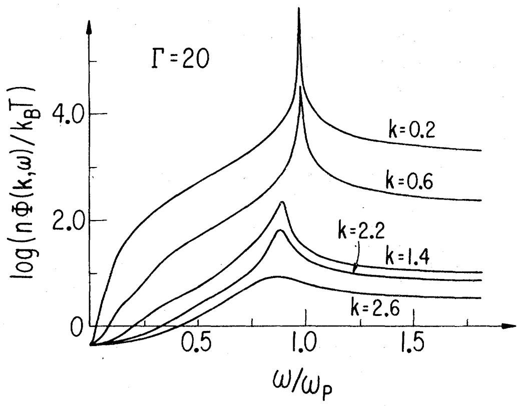

FIG. 2. Effective frequency and wave-vector-dependent interaction $\Phi(k,\omega)$ for $\Gamma = 20$ as a function of ω/ω_p for various wave vectors $q = ka$. The logarithm of $n\Phi(k,\omega)/k_B T$ is shown because of the rapid variation of $\Phi(k,\omega)$ near $\omega = \omega_p$ for $k \lesssim 2$.

The dimensionless "frequency" ν and "wave vector" k are given by $\nu = (3\Gamma/2)^{1/2}(\omega/\omega_p k)$ and $k = qa$, where $a = (3/4\pi n)^{1/3}$. For large ν, $2\nu F(\nu) \to 1 + 1/2\nu^2$ and for small k, $S(k) \to k^2/3\Gamma$. Thus from (5.5) and (5.6) it is easy to see that in the limit of small k, $\epsilon(k,\nu)$ has a zero for $\nu = (3\Gamma/2)^{1/2}k^{-1}$ or $\omega = \omega_p$. The zero in ϵ and pole in Φ at $\omega = \omega_p$ corresponds to the well-known[4] collective plasma oscillations. The frequency and wave-vector dependence of $\Phi(q,\omega)$ is evaluated numerically and shown in Fig. 2 for $\Gamma = 20$. It is seen that the plasma oscillations are well defined for $k \lesssim 2$.

B. Effective-interaction approximation for D

The self-diffusion constant D can be directly related to the time integral of the memory function $M(t)$ of the velocity autocorrelation function by[9]

$$D^{-1} = V_0^{-2} \int_0^{+\infty} dt\, M(t)\,. \tag{5.7}$$

The relation (5.7) is exact and follows from (1.3) and (1.4). We will calculate D in the one Sonine-polynomial approximation (see Appendix B) by substituting $M_{1p}(t)$ in (5.7). This approximation will lead to qualitative results for D. We reserve an examination of higher-order polynomial solutions for D to future work. It is convenient to rewrite (5.7) in dimensionless units and introduce the dimensionless quantities $D^* = D/\omega_p a^2$, $\tau = t\omega_p$, and $\bar{M} = M/\omega_p^2$, so that (5.7) becomes

$$D^{*-1} = 3\Gamma \int_0^{+\infty} d\tau\, \bar{M}(\tau)\,. \tag{5.8}$$

It is more direct to obtain $\bar{M}_{1p}^{\rm EIA}(\tau)$ from (3.10) rather than from (4.11) and (3.8). S_s is found from (2.7), (4.1), and the general relation between the Laplace and Fourier transform and has the Gaussian form

$$S_s(k,\tau)=e^{-k^2\tau^2/6\Gamma}\,. \tag{5.9}$$

The "mean-field" form of G is found from (4.9) and (5.5) and is given by

$$G(k,\tau)=\frac{2S(k)}{\sqrt{\pi}}\int_0^{+\infty}d\nu\,\frac{e^{-\nu^2}\cos[k\nu\tau(2/3\Gamma)^{1/2}]}{D^2(k,\nu)}\,. \tag{5.10}$$

In (5.10) we have used the fact that $D(k,\nu)=D(k,-\nu)$. The form (5.10) for G implies that $G(k,\tau)$ reduces to the free particle form (5.9) for $k\gg 1$, since in this limit $S(k)$ and $D(k,\nu)$ approach unity. If we express (5.10) as a complex integral, it is easy to establish that $G(k,\tau=0)=1$ as expected, and $G(k=0,t)=\cos\omega_p t$. This oscillatory behavior of G for small k is a consequence of the collective plasma oscillations. For k corresponding to $S(k)\leq 0.1$, the integrand in (5.10) is a rapidly varying function of ν and difficult to treat numerically. We are led to adopt the approximation that for $k<k_0$

$$G(k,\tau)=\cos\omega_k\tau e^{-k^2\tau^2/6\Gamma}\quad(k<k_0)\,, \tag{5.11}$$

and

$$\omega_k^2=k^2/3\Gamma S(k)\,, \tag{5.12}$$

where k_0 is determined by the condition that $S(k_0)=0.1$. Note that k_0 is an increasing function of Γ as is indicated by the small-k behavior of $S(k)=k^2/3\Gamma$ [see (4.6)]. For $k>k_0$, $G(k,\tau)$ can be obtained from (5.10) by standard numerical methods. The dimensionless "dispersion relation" ω_k, (5.12), is determined from the real part of the zero of $\epsilon(k,\nu)$ for small k and reduces to unity for $k=0$. The dispersion relation also has a small imaginary part which leads to the damping of the plasma oscillations. Since the contribution of $G(k,\tau)$ for $k<k_0$ to D will prove to be relatively unimportant, we have chosen in (5.10) the damping factor to be of the free-particle form $e^{-k^2\tau^2/6\Gamma}$. The advantage of the Gaussian form is that it will allow us to make a simple interpolation formula between (5.11) and the hydrodynamic limit (4.14).

The presence for small k of the oscillatory $\cos\omega_k\tau$ factor in the mean-field form for G leads to large cancellations in the time integral of $\overline{M}(\tau)$. The absence of the plasma oscillations in the free-particle form for G [see (4.2) or (5.9)] indicates why M^{EIA} and M^{FP} yield large quantitative differences in the calculated vlaues of D. It is also expected that the contribution to D^* for $k<k_0$ should be small compared to that for $k>k_0$.

An explicit form [see (5.21)] of M_{1p}^{EIA} can be found from (3.10) and (5.9)-(5.11). The large-t behavior of $M_{1p}^{\mathrm{EIA}}(t)$ is easily determined to be

$$M_{1p}^{\mathrm{EIA}}(t)\rightarrow\tfrac{1}{3}e^2(2m\beta^3/\pi)^{1/2}t^{-3}\cos\omega_p t\,, \tag{5.13}$$

which should be compared with (4.8) and (4.17).

To calculate D we use (5.8) and the explicit form of $\overline{M}_{1p}^{\mathrm{EIA}}(\tau)$ discussed above, perform the τ integral analytically, and write the zeroth-order approximation to D in the one-Sonine-polynomial approximation as

$$D_{1p}^{*(0)}\equiv D_0^*=(27\pi/\Gamma)^{1/2}(I_1+I_2)^{-1}\,, \tag{5.14}$$

where

$$I_1=\int_0^{k_0}dk\,k^3\{[S(k)-1]^2/S(k)\}e^{-1/4S(k)} \tag{5.15}$$

and

$$I_2=\left(\frac{8}{\pi}\right)^{1/2}\int_{k_0}^{\infty}[S(k)-1]^2\int_0^{\infty}\frac{d\nu\,e^{-2\nu^2}}{D^2(k,\nu)}\,. \tag{5.16}$$

In I_1 and I_2 we have used (2.15), (2.18), and (2.19) to express $C_D(k)$ in terms of $S(k)$. I_1 and I_2 are evaluated numerically using as input the "exact" Monte Carlo computations[14] of $S(k)$. The values $\Gamma=4, 20$, and 110 are considered, for which the "cut off" parameter k_0 is found to be 1.0, 1.8, and 2.8, respectively. The results for I_1, I_2, and D_0^* are given in Table I and compared to the molecular dynamics values D_{MD}^*. Note that $I_1\ll I_2$ as expected. We see that the calculated values of D_0^* exhibit a rapid variation in Γ consistent with the behavior of D_{MD}^* and that the ratio D_0^*/D_{MD} is approximately 1.5 for $\Gamma=4$ and increases to approximately 2 at $\Gamma=110$. The slow convergence of the Sonine-polynomial expansion for the calculation of the transport properties of a plasma is known[26] for $\Gamma\ll 1$ and in this limit causes the one Sonine-polynomial calculation of D to differ from the exact result by a factor of approximately 1.5. The nature of the convergence of the Sonine-polynomial expansion for arbitrary Γ is not known. We conjecture that the slow convergence will persist and that the additional discrepancy between D_{MD}^* and $2D_0^*/3$ for $\Gamma=110$ mainly arises from the error in $M^{\mathrm{EIA}}(t)$ for very short times due to the incorrect

TABLE I. Comparison of molecular-dynamics and microscopic calculations of the dimensionless self-diffusion constant D^* as a function of Γ. The integrals I_1 and I_2 are defined in (5.15) and (5.16) respectively and are related to D_0^* by (5.14).

Γ	I_1	I_2	D_0^*	D_{MD}^*
4	0.146	6.53	0.69	0.46
19.7	1.18	21.1	0.092	0.060
110.4	2.97	94.9	0.0090	0.0051

treatment of close collisions within the disconnected approximation. The additional contribution to D from the inclusion of the hydrodynamic modes important in the long-time behavior of $M(t)$ is briefly discussed in Sec. V C. The qualitative agreement between D_0^* and D_{MD}^* leads to the conclusion that the effective-interaction approximation yields a good zeroth-order approximation for the self-diffusion constant.

C. Interpolation formula for $M(t)$

We seek a first-order approximation $M_{1p}^{(1)}(t)$ by approximating the intermediate correlation functions S_s and G using forms which interpolate between their mean-field behavior (5.9) and (5.10) and their long-time hydrodynamic behavior (4.13) and (4.14). In the spirit of our iterative approach $M_{1p}^{(1)}(t)$ will be expressed in terms of the transport coefficients obtained from the zeroth-order approximation. It is straightforward to choose a simple interpolation form for S_s, since it has the same Gaussian q dependence in both (5.9) and (4.13). We adopt the form[10]

$$S_s(k,\tau)=\exp[-k^2W_D(\tau)/2]\,, \tag{5.17}$$

$$W_D(\tau)=2D_0^*\{\tau+3\Gamma D_0^*[\exp(-\tau/3\Gamma D_0^*)-1]\}. \tag{5.18}$$

Note that W_D is expressed in terms of D_0^* determined in the effective-interaction approximation. However, since D_0^* was obtained in Sec. V B in the one-Sonine-polynomial approximation, we take D_0^* in (5.18) to be the values shown in Table I corrected by an overall factor of $\frac{2}{3}$. The choice of an interpolation formula for G is more complicated because of the presence of a $\cos\omega_p t$ factor in both (4.14) and (5.11). We make a simple modification of (5.11) so that $G(k,\tau)$ includes the hydrodynamic modes for $k<k_0$:

$$G(k,\tau)=\cos\omega_k\tau\exp(-k^2W_{nn}(\tau)/2)\quad(k<k_0)\,, \tag{5.19}$$

$$W_{nn}(\tau)=\tfrac{4}{3}\eta^*[\tau+2\eta^*\Gamma(e^{-\tau/2\Gamma\eta^*}-1)]. \tag{5.20}$$

For $k>k_0$, $G(k,\tau)$ is given by (5.10). In (5.20) we have neglected the bulk viscosity and considered only the shear-viscosity contribution to Γ_{nn} [see (4.15)]. The dimensionless viscosity $\eta^*=\eta/nm\,\omega_p a^2$. A zero-order calculation of η^* would require the effective-interaction-approximation form for $\phi^{(c)}$. We anticipate that such a calculation of η^* would lead to reasonable values. Since we have not as yet performed such a microscopic calculation, we adopt the results for η^* obtained from a generalized hydrodynamics calculation.[27] The qualitative features of this calculation are that η^* decreases rapidly with increasing Γ for $\Gamma<10$, passes through a minimum around $\Gamma=20$, and then increases slowly with increasing Γ. For $\Gamma=4$, 20, and 110 the numerical results are $\eta^*=0.12$, 0.078, and 0.12, respectively.

We combine (3.10), (5.17), (5.19), and (5.10) and write $\overline{M}_{1p}^{(1)}(\tau)$ in the form

$$\overline{M}_{1p}^{(1)}(\tau)=(2/27\pi\Gamma)[M_1(\tau)+M_2(\tau)]\,, \tag{5.21}$$

$$M_1(\tau)=\int_0^{k_0}dk\,k^4\{[S(k)-1]^2/S(k)\}\cos\omega_k\tau \times\exp\{-k^2[W_D(\tau)+W_{nn}(\tau)]/2\}\,, \tag{5.22}$$

$$M_2(\tau)=\frac{2}{\sqrt{\pi}}\int_{k_0}^{\infty}dk\,k^4[S(k)-1]^2\exp[-k^2W_D(\tau)/2] \times\int_0^{\infty}d\nu\,e^{-\nu^2} \times\cos[k\tau(2/3\Gamma)^{1/2}\nu]D^{-2}(k,\nu)\,. \tag{5.23}$$

Note that if we set $W_D(\tau)=W_{nn}(\tau)=k^2\tau^2/6\Gamma$ in (5.22) and (5.23), the form of $\overline{M}_{1p}^{(1)}(\tau)$ reduces to $\overline{M}_{1p}^{\mathrm{EIA}}(\tau)$. It is necessary to include the hydrodynamic modes in (5.21) in order to obtain a realistic approximation for $\overline{M}(\tau)$ in the range $0\lesssim\tau\lesssim20$ considered in the MD calculation. For example, an inspection of (5.18) and (5.21) shows that the hydrodynamic diffusive mode contributes to $\overline{M}^{(1)}(\tau)$ for $\tau>\tau_D\sim3\Gamma D^*$, which ranges from $O(5)$ to $O(1)$ in the range of Γ considered. The optimal determination of a cutoff parameter k_0 and a better interpolation formula for G as well as S_s requires further investigation. We expect however that we have described G and S_s in the hydrodynamic region ($k\ll1$, $\tau\gg1$) and the free-particle region ($\tau\ll1$ or $k\gg1$) accurately. It is assumed that G is well represented by the effective interaction approximation in the intermediate τ range for $k>k_0$ and by

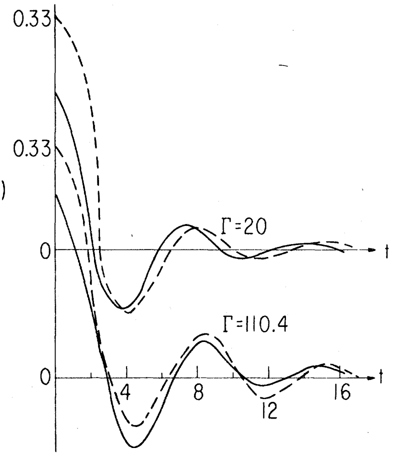

FIG. 3. Comparison of memory function obtained from molecular dynamics (dashed line, Ref. 7) and from the microscopic theory [solid line, Eq. (5.21)] for $\Gamma=19.7$ and 110.4. The time scale is in units of ω_p^{-1} and $M(t)$ is in units of ω_p^{-2}.

the interpolation formula (5.19) for $k<k_0$. S_s and G are expected to be slowly varying functions in the intermediate space-time region.

The interpolation formula (5.21) for $\bar{M}_{1p}^{(1)}(\tau)$ is evaluated numerically using the Monte Carlo computation[14] of $S(k)$ and the numerical values of D^* and η^* as indicated above. The results for $\Gamma = 4, 20$, and 110 are compared to the exact MD results[7] for $M(\tau)$ in Fig. 3. It is seen that the calculated $\bar{M}_{1p}^{(1)}(\tau)$ is in qualitative agreement with the MD values for $\bar{M}(\tau)$ except for τ near zero, for which the basic approximation (3.1) breaks down.

In principle we should use the $\bar{M}^{(1)}(\tau)$ to obtain the first-order correction to D_0^* which would arise from the inclusion of the hydrodynamic modes. However since we have calculated D_0^* only in the first-Sonine-polynomial approximation and have not performed a microscopic calculation of η_0^* we have not yet performed such an investigation.

VI. DISCUSSION

It has been shown that we can find relatively simple approximations that lead to a reasonable description of the dynamical properties of an equilibrium classical one-component plasma for which there is no intrinsic small parameter. Many simplifications were made in the analysis in order to make clear the essential features of the method. In this section we discuss the qualitative features of our results and indicate areas of future work.

A striking feature of our calculation and of the MD results is that for large Γ the single-particle motion is dominated by its coupling to the collective plasma oscillations. The dominance of the collective oscillations at high density for a wide range of k and ω can also be seen from the MD results[7] for the dynamic structure function $S(k, \omega)$. An essential difference between an OCP and a neutral fluid, with short-range interactions, is that the collective plasma oscillations dominate the long-wavelength response of an OCP in both the short-time (mean field) and the long-time (hydrodynamic) limits in contrast to the collective sound mode in a neutral fluid, which is important in only the hydrodynamic limit. Another consequence of the long-range nature of the Coulomb potential is that the frequency of the dispersion relation of the plasma oscillations in both the long-time and short-time description of an OCP is identical and finite in the long-wavelength limit. It is easy to see that the collective properties of an OCP extend over a wider range of k and ω as Γ increases. For short times or high frequencies ($\omega \gg \omega_p$) collisions are not important, and the mean-field description of $S(k, \omega)$ is valid. In this case we found in Sec. V B that the plasma mode is well defined for $k \leq k_0$. Since k_0 is an increasing function of Γ, the collective properties of an OCP for $\omega \gg \omega_p$ extend over a wider range of k as Γ increases. The hydrodynamic description is valid for $k<1$ and for times greater than the mean time between collisions. Since a measure of this time is D, which is a decreasing function of Γ, the hydrodynamic description extends over a larger range of time as Γ increases.

We have seen that the fully renormalized kinetic-theory approach[9-11] is a flexible formalism which can be successfully applied to both dense plasmas and dense neutral fluids. The basic features of the formalism are the introduction of a memory function in a generalized kinetic equation to describe the effects of the interactions, the expression of the memory functions in terms of an effective two-body problem, and the systematic inclusion of the exact static correlation functions. For the case of a plasma a simple approximation, (3.1), which takes into account the long-range correlations in the system was introduced for the dynamical evolution of two particles in the medium. This simple approximation is part of a systematic and well-defined approximation procedure in which the terms that have been neglected can be analyzed and estimated.

The calculations presented here of the memory function of the velocity auto correlation functions and of the self-diffusions constant have been relatively crude and the results qualitative. However, the calculation lead to reasonable agreement with the molecular-dynamics results, illustrate the nature and level of complexity of microscopic calculations for a dense plasma, and indicate areas where further work is needed and additional applications are possible. The present calculations can be made quantitative if we use the method of kinetic modeling[19] (described briefly in Appendix A) to obtain a quantitative solution of the generalized kinetic equation (2.24), and if we correctly treat the close collisions. Neither extension of the present calculation poses any essential difficulty.

It would be of interest to investigate the van Hove self-correlation function, $S_s(k, \omega)$, since MD results[28] for S_s are expected to become available in the near future. Such a calculation would involve the calculations of a k-dependent memory function and thus, in general, the numerical evaluation of an angular integral.

Although we have only considered the motion of a tagged particle in this paper, the approach can be applied to the study of the phase-space density correlation function (2.9) and its associated memory function (2.25). Such an investigation would yield numerical results as a function of Γ for the transport coefficients (e.g., the shear and bulk viscosities and thermal conductivity), and numerical results for the dynamic longitudinal and transverse current correlation functions as a function of Γ, k, and ω. These calculations would be of much interest since the available MD results would serve as a test of the microscopic theory, which could be extended to yield new information about the dynamics of the OCP. It is also straightforward to extend the FRKT formalism to multicomponent systems[29] as well as to quantum systems. It would be of interest to investigate the behavior of the transport coefficients, in particular the thermal conductivity as a function of density and temperature of a laser-compressed plasma in which quantum effects are important. Such a plasma corresponds to the case[2] in which the positive ions are classical and characterized by a Γ of 2–5 and the electrons are highly degenerate. We hope to consider these problems in future work.

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APPENDIX A

We discuss briefly the assumptions underlying the relation (3.8) between $M_{1p}(t)$ and $\varphi_s(k, pp', t)$ and outline the method of kinetic modeling.[19] To simplify the discussion we introduce the dimensionless momentum variable $\xi = p/mV_0$ and the dimensionless weight function

$$W_0(\xi) = (2\pi)^{-3/2} e^{-\xi^2/2} .$$

We introduce a set of basis functions $\psi_\alpha(\xi)$ which satisfy the orthogonality and completeness conditions

$$\int d^3\xi \, W_0(\xi)\psi_\alpha(\xi)\psi_\beta(\xi) = \delta_{\alpha,\beta} , \qquad \text{(A1)}$$

$$\sum_{\alpha=1}^{\infty} \psi_\alpha(\xi)\psi_\alpha(\xi') W_0(\xi) = \delta(\xi - \xi') , \qquad \text{(A2)}$$

and write φ_s as

$$\varphi_s(k, \xi\xi'.z) = \sum_{\alpha,\beta} \psi_\alpha(\xi)\psi_\beta(\xi') W_0(\xi)\varphi_{\alpha\beta}(k,z) . \qquad \text{(A3)}$$

The matrix elements $\varphi_{\alpha\beta}(k,z)$ are defined by

$$\varphi_{\alpha\beta}(k,z) = \int d^3\xi d^3\xi' \, \psi_\alpha(\xi)\psi_\beta(\xi')\varphi_s(k, \xi\xi', z) W_0(\xi') . \qquad \text{(A4)}$$

Although we are interested in the limit $k = 0$ [see (2.8)], we consider the general case of finite k, for which φ_s does not have rotational symmetry. In this case the usual Sonine polynomials[30] are no longer an appropriate choice for the basis functions, and we choose the functions

$$\psi_{lmn}(\xi) = (l!\,m!\,n!)^{-1/2} \bar{H}_l(\xi_x)\bar{H}_m(\xi_y)\bar{H}_n(\xi_z) , \qquad \text{(A5)}$$

where

$$\bar{H}_l(\xi) = 2^{-l/2} H_l(\xi/\sqrt{2}) . \qquad \text{(A6)}$$

H_l is the usual Hermite polynomial.[31] The calculation of the matrix elements $\varphi_{\alpha\beta}$ is one of the major tasks of kinetic modeling.

The approximate φ_s, (3.7), satisfies the conservation of particles condition

$$\int d^3\xi \, \varphi_s(k, \xi\xi', z) = 0 ,$$

but does not satisfy conservation of momentum and energy since the tagged particle can exchange momentum and energy with the medium. To model φ_s in (A3) we isolate the hydrodynamic state $\psi_1 \equiv \psi_{000} = 1$ associated with number conservation, assume that the off-diagonal matrix elements are small compared to the diagonal matrix elements, and for $\alpha > 2$ approximate the diagonal matrix elements by $\varphi_{22} \equiv \varphi_{001,001}$, the first nonhydrodynamic matrix element. (Recall that $\psi_2 \equiv \psi_{001} = \xi_z$.) With these assumptions and the use of (A2) we write (A3) as

$$\varphi_s(k, \xi\xi', z) = -\varphi_{22}(k,z) W_0(\xi) + \varphi_{22}(k,z)\delta(\xi - \xi') . \qquad \text{(A7)}$$

In general we construct a kinetic model of order N by assuming that $\varphi_{\alpha\beta} = \varphi_{NN}\delta_{\alpha\beta}$ for all $\alpha, \beta > N$. The substitution of the order-N generalization of (A7) into (2.24) yields a matrix equation for

$$C_{\alpha\beta}(k,z) = \int d^3\xi \, d^3\xi' \, \psi_\alpha(\xi)\psi_\beta(\xi') C_s(k, \xi\xi', z) . \qquad \text{(A8)}$$

The solution of this matrix equation for N large is the second major task of kinetic modeling.

To solve for $V_D(t)$ we use the general relations

$$\begin{aligned} V_D(z) &= V_0^2[z - M(z)]^{-1} \\ &= \lim_{k\to 0} \frac{V_0^2}{n} C_{22}(k,z) , \end{aligned} \qquad \text{(A9)}$$

where $V_D(z)$ and $M(z)$ are the Laplace transforms of $V_D(t)$ and $M(t)$ defined by (1.2) and (1.4), respectively. The substitution of the approximation

(A7) into (2.24) yields

$$\frac{1}{n}C_{22}(k,z) = \frac{\Lambda_{22}(k,z)+\varphi_{22}(k,z)[\Lambda_{11}(k,z)\Lambda_{22}(k,z)-\Lambda_{12}^2(k,z)]}{1+\varphi_{22}(k,z)\Lambda_{11}(k,z)}, \tag{A10}$$

where

$$\Lambda_{\alpha\beta}(k,z) = \int d^3\xi \frac{\psi_\alpha(\xi)\psi_\beta(\xi)W_0(\xi)}{z - V_0\vec{k}\cdot\vec{\xi} - \varphi_{22}(k,z)}. \tag{A11}$$

In the limit $k\to 0$, (A10) reduces to the form given by the usual one-Sonine-polynomial expansion and the approximate relation (3.8) follows. We plan to consider in future work the contribution of higher-order matrix elements to C_{22}.

APPENDIX B

The relation between the long-time limit of $M(t)$ and $V_D(t)$ for the case where their long-time behavior is nonexponential has been given in Ref. 32. We apply these results to the case where for $t\to\infty$; $M(t)$ is given by [see (4.16)]

$$M(t)\to Ct^{-3/2}\cos\omega_p t \tag{B1}$$

where C is a constant. We will show that for $t\to\infty$

$$V_D(t)\to - V_0^2(2\pi\omega_p C)^{-1}t^{-3/2}\sin\omega_p t. \tag{B2}$$

To make contact with the notation of Ref. 32, we define the Laplace transform $\tilde{V}_D(s)$ as

$$\tilde{V}_D(s) = \int_0^{+\infty} dt\, e^{-st}V_D(t). \tag{B3}$$

The long-time behavior of $V_D(t)$ and $M(t)$ will be extracted from the behavior of $\tilde{V}_D(s)$ and $\tilde{M}(s)$ in the neighborhood of their dominant singularities. For the case (B2) the $t\to\infty$ behavior of $V_D(t)$ is regulated by a pair of branch points lying at $s=\pm i\omega_p$. We have

$$\tilde{V}_D(s) = -iV_0^2(2\pi^{1/2}\omega_p C)^{-1}[(s-i\omega_p)^{1/2}-(s+i\omega_p)^{1/2}] \tag{B4}$$

for s near the branch points. In this case $\tilde{V}_D(s)$ approaches a unique, finite limit as $s\to\pm i\omega_p$ along any direction in the half-plane $\mathrm{Re}\,s>\epsilon<0$. Under this condition, which is a slight generalization of the condition that the diffusion constant exists, it can be shown[32] that for $t\to\infty$

$$V_D(t)\to -(1/\pi)\,\mathrm{Im}\{e^{i\omega_p t}X(t)\}, \tag{B5}$$

$$M(t)\to (V_0^2/\pi)\,\mathrm{Im}\{e^{i\omega_p t}X(t)/\tilde{V}^2(i\omega_p)\}, \tag{B6}$$

where

$$X(t) = \int_0^{+\infty} d\eta\, e^{-\eta t}[\tilde{V}_+(\eta) - \tilde{V}_-(\eta)] \tag{B7}$$

and

$$\tilde{V}_\pm(\eta) = \tilde{V}_D(i\omega_p + \eta e^{\pm i\pi}). \tag{B8}$$

We first apply the above to the more familiar case where $V_D(t)\to t^{-3/2}$, i.e., $\omega_p=0$. Then since $\tilde{V}_D(0)=D$, we have $V_D(t)\to -(mD/k_B T)^2 M(t)$ for $t\to\infty$. In our case $X(t)\to V_0^2(2\omega_p C t^{3/2})^{-1}$ and $\tilde{V}_D^2(i\omega_p) = -iV_0^4(2\pi\omega_p C^2)^{-1}$, and the result (B1) follows from (B2), (B5), and (B6). Note that in the present case $V_D(t)$ and $M(t)$ differ in phase as well as amplitude.

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