Coupling of Single-Particle and Collective Motions in a One-Component Plasma

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We perform a microscopic calculation of the memory function of the velocity autocorrelation function in a classical one-component plasma. Our theory yields oscillatory behavior in the velocity autocorrelation function consistent with recent molecular-dynamics computations. We also calculate the self-diffusion coefficient as a function of density.

Recently Hansen, Pollock, and McDonald[1, 2] have reported molecular-dynamics (MD) computations of the velocity autocorrelation function $V_D(t)$ and its memory function $M(t)$ for the classical one-component plasma in a uniform background of opposite charge. The most striking feature of their computations was a coupling of the single-particle motion to the collective density fluctuations. This coupling was manifested in the appearance at high densities and at long times 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 here a simple microscopic theory which gives the oscillatory feature of $V_D(t)$ as a function of the dimensionless parameter[3] $\Gamma = e^2/ak_BT$ with $a = (3/4\pi n)^{1/3}$. We calculate, as functions of Γ, $M(t)$ and the time integral of $M(t)$ which is related to the self-diffusion constant D, and we find qualitative agreement with the MD computations.

The microscopic theory is based on the kinetic theory formalism presented by Mazenko[4-6] (whose notation we adopt) and leads to a kinetic equation that includes the exact statics of the system, treats screening at large and small distances consistently, and is valid for all wave vectors k and frequencies ω. In the limit of small k and ω and $\Gamma \ll 1$ the kinetic equation reduces to the Lenard-Balescu-Guernsey (LBG) equation.[7] Our kinetic equation is free of the short-range divergence due to close collisions associated with the LBG equation.

The velocity autocorrelation function is defined as

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

and can be obtained from[4] the time-dependent phase-space correlation function for self-diffusion,

$$C_s(12, t) = \langle \delta f_s(2) e^{+iLt} \delta f_s(1)\rangle . \tag{2}$$

$\delta f_s(1)$ is the deviation of the phase-space density for a tagged particle,

$$f_s(1) = \sqrt{N}\,\delta(1 - q_1), \tag{3}$$

from its equilibrium value and $q_1 = (r_1, p_1)$ are the phase-space coordinates of particle 1. We denote the Liouville operator by L and the average over an equilibrium ensemble by $\langle\,\rangle$. We work mostly with the temporal Laplace and spatial Fourier transform $C_s(k, p_1p_2, z)$. We also require the time-dependent phase-space density correlation function $C(12; t)$ defined as in (2) with $f_s(1)$ replaced by

$$f(1) = \sum_{j=1}^{N} \delta(1 - q_j).$$

The memory function φ_s is introduced as the kernel in the kinetic equation for C_s,

$$\left(z - \frac{k\cdot p_1}{m}\right) C_s(k, p_1p_2, z) - \int d^3\bar{p}\, \varphi_s(k, p_1\bar{p}, z) C_s(k, \bar{p}p_2, z) = \tilde{C}_s(k, p_1p_2), \tag{4}$$

where $\tilde{C}_s(k, p_1p_2) = C_s(k, p_1p_2, t=0)$. The calculation of φ_s is the major problem in developing a microscopic theory. We use the analysis of Ref. 6 to write φ_s in the exact form [see Eqs. (5.8) and (5.15) of Ref. 6]

$$\varphi_s(12) n f_0(p_2) = -\int d\bar{1}\, d\bar{2}\, d\bar{3}\, d\bar{4}\, \mathcal{V}_s(1; \bar{1}\bar{3}) \bar{G}_s(\bar{1}\bar{3}; \bar{2}\bar{4}) \mathrm{V}_s(2; \bar{2}\bar{4}) . \tag{5}$$

The renormalized interaction $\mathcal{V}_s$ is determined by the two-particle statics, $\overline{G}_s$ is a phase-space correlation function that describes the dynamical evolution of two particles in the medium, and $f_0(p)$ is the renormalized Maxwellian distribution.

We assume that the dominant physical processes contributing to $\overline{G}_s$ are the collective effects associated with the long-range nature of the Coulomb potential and retain those events in which two particles initially interact, propagate independently of each other but interact with the medium, and eventually recollide. The correlations that lead to the recollision are "communicated" by the medium, and hence the process is associated with collective effects. We thus approximate $\overline{G}_s$ by its disconnected part

$$\overline{G}_D(11';22';t)=\int d\overline{1}\,d\overline{2}\,d\overline{3}\,d\overline{4}\,\tilde{C}_s^{-1}(1\overline{1})\tilde{C}^{-1}(2\overline{2})\tilde{C}_s^{-1}(1'\overline{4})\tilde{C}^{-1}(2'\overline{3})C_s(\overline{1}\overline{4};t)C(\overline{2}\overline{3};t). \tag{6}$$

The "disconnected" approximation (6) ignores the effects of close collisions and can be expected to break down for very short times. This approximation can be made part of a systematic analysis in which the remaining contributions to φ can be explicitly calculated. We will discuss the contribution of close collisions in future work. It is straightforward to show that if we substitute (6) into (5), we find

$$\varphi_s(k,p_1p_2,t)nf_0(p_2)=-\int d^3q(2\pi)^{-3}[\beta^{-1}C_D(q)]^2\vec{q}\cdot\nabla_{p_1}\vec{q}\cdot\nabla_{p_2}C_s(k-q,p_1p_2,t)S_{nn}(q,t), \tag{7}$$

where

$$S_{nn}(q,t)=\int d^3p_3\,d^3p_4C(q,p_3p_4,t) \tag{8}$$

is the usual time-dependent density-density correlation function and $C_D(q)$ is the direct correlation function.

Given (7) and (4) we must solve a nonlinear integral equation in the momentum variables. To avoid this technical difficulty, which can be treated by use of the method of kinetic modeling,[8] we present an approximate solution which gives the quantitative long-time behavior and the qualitative short-time behavior. We assume that the correct long-time behavior is dominated by the single hydrodynamic mode associated with number conservation:

$$C_s(k,p_1p_2,t)\approx f_0(p_1)f_0(p_2)S_s(k,t). \tag{9}$$

Equation (9) leads directly to the memory function

$$M(t)=[3(mV_0)^2n]^{-1}\int d^3q\,(2\pi)^{-3}q^2[\beta^{-1}C_D(q)]^2S_s(q,t)S_{nn}(q,t), \tag{10}$$

where $S_s(q,t)$ is defined as in (8) with C replaced by C_s, and $V_0^2=(\beta m)^{-1}$. The approximate solution (10) for $M(t)$ is equivalent to the first Sonine-polynomial approximation used in treating the Boltzmann equation.

In principle we should use the self-consistent solution of (4) and (7) for S_s and the solution of a similar set of equations for S_{nn} in (10) to determine $M(t)$. In lieu of such a sophisticated approach we start with the simplest possible forms for S_s and S_{nn} and identify those features of S_s and S_{nn} that make significant contributions to $M(t)$ and D. We calculate $M(t)$ by approximating S_s and S_{nn} in (10) in three ways: (a) by their free-particle forms, (b) by mean-field theory, and (c) by using forms which interpolate between the exact short-time and long-time hydrodynamic behavior.

(a) The free-particle forms for S_s and S_{nn} yield results for D which are off by two orders of magnitude from the MD calculations.

(b) In the mean-field approximation we neglect the collisional parts of the kinetic equations for C and C_s and retain only the static contribution to φ. (The static contribution to φ_s vanishes identically.) These approximations for C_s and C can be solved exactly to give the free-particle form

$$S_s(q,t)=n\exp(-q^2V_0^2t^2/2) \tag{11}$$

and the "self-consistent-field" result

$$S_{nn}(q,t)=-i\int dze^{-izt}\frac{nS(q)Q(q,z)}{\epsilon(q,z)}, \tag{12}$$

where

$$\epsilon(q,z)=\{1+[S(q)-1]zQ(q,z)\}/S(q), \tag{13}$$

$$Q(q,z)=\int d^3p\,f_0(p)(z-\vec{q}\cdot\vec{p}/m)^{-1}. \tag{14}$$

The substitution of (11) and (12) in (10) yields a nontrivial form $M_0(t)$ which reduces to the LBG form in the appropriate limits. We choose $M_0(t)$ as our zeroth-order approximation from which we calculate D_0. We use the MD computation[9] of $S(q)$, the static structure function, as input and perform the integrals numerically. The results are given in terms of the dimensionless quantity $D^* = D/\omega_p a^2$ and are listed in Table I. The calculated $D_0{}^*$ exhibits a rapid variation in Γ consistent with the MD calculations[2]; for $\Gamma = 4$ and 20, $D_0{}^*/D_{\rm MD}{}^* \sim 1.5$, and for $\Gamma = 110.45$, $D_0{}^*/D_{\rm MD}{}^* \sim 1.8$. A simple calculation of D^* for $\Gamma < 1$, for which the contribution of close collisions is negligible, shows that the use of (9) causes $D_0{}^*$ to be too large by a factor of approximately 1.5. Hence, the additional discrepancy between $D_0{}^*$ and $D_{\rm MD}{}^*$ for $\Gamma = 110.45$ is due to the use of the disconnected approximation. The significant difference between the free-particle approximation for S_{nn} and (12) is that for small q Eq. (12) is proportional to a factor $\cos(\omega_p t)$ whose oscillations lead to large cancelations in the integral over $M(t)$ and cause the large quantitative difference in the calculated values of D.

(c) Although M_0 is a useful approximation and gives reasonable values for D it does not give the long-time behavior of $M(t)$ accurately. This is because the mean-field approximations for S_s and S_{nn} neglect the hydrodynamical modes that dominate the long-time behavior of $M(t)$. The interpolation forms[4] mentioned above are

$$S_s(q,t) = n\exp[-q^2 W_D(t)/2], \tag{15}$$

$$W_D(t) = 2D\left\{t + \frac{D}{V_0{}^2}\left[\exp\left(\frac{-V_0{}^2 t}{D}\right) - 1\right]\right\}. \tag{16}$$

For $q < q_0$ we choose

$$S_{nn}(q,t) = nS(q)\cos(\omega_q t)\exp[-q^2 W_{\Gamma_{nn}}(t)/2], \tag{17}$$

where $\omega_q{}^2 = \omega_p{}^2 q^2/4\pi n e^2 S(q)\beta$ and $W_{\Gamma_{nn}}(t)$ is defined as in (16) except that D is replaced by the sound attenuation coefficient[10] Γ_{nn}. For $q > q_0$ $S_{nn}(q,t)$ is given by (12). The value of q_0 is determined by the criterion $S(q_0) = 0.1$ which implies that q_0 is an increasing function of Γ. Equations (15)–(17) require D and Γ_{nn} as functions of Γ. Since our calculated D_0 is off by an overall factor, we have substituted the MD values of D in (16). Also since we have not calculated Γ_{nn}, we use the values determined from a generalized hydrodynamic calculation.[11]

The interpolation formula for $M(t)$ is evaluated numerically and compared to the MD data for $M(t)$ in Fig. 1. It is seen that the calculated $M(t)$ is in qualitative agreement with $M_{\rm MD}(t)$. In the long-time limit the dominant contribution to the integral in (10) arises from small q for which $S(q)$ and $C_D(q)$ are given exactly by their Debye-Hückel forms,[2] and the dynamics is given exactly by hydrodynamics. We thus obtain the asymptotic behavior of $M(t)$,

$$M(t) \to \frac{\sqrt{2}\,e^2}{3\pi n(D + \Gamma_{nn}/2)}\, t^{-3/2}\cos(\omega_p t). \tag{18}$$

Equation (18) depends[12] only on (6) and (9). We see that for long times the motion of single particles is coupled to the wavelength density fluctuations and leads to a long-time tail which oscillates about zero with a frequency ω_p in qualitative agreement with the MD calculation of $V_D(t)$ and the "exact" $M(t)$. Since we have not calculated Γ_{nn} we have not used the interpolation form for $M(t)$ to obtain the correction to $D_0{}^*$ which arises from the inclusion of the hydrodynamic modes.

Our results indicate that the disconnected approximation (6) gives a good description of the long-time and intermediate-time behavior of the

TABLE I. Comparison of molecular-dynamics and microscopic calculations of the dimensionless diffusion constant D^* as a function of Γ.

Γ	$D_{\rm MD}{}^*$	$D_0{}^*$
4	0.46	0.69
20	0.060	0.092
110.45	0.0051	0.0090

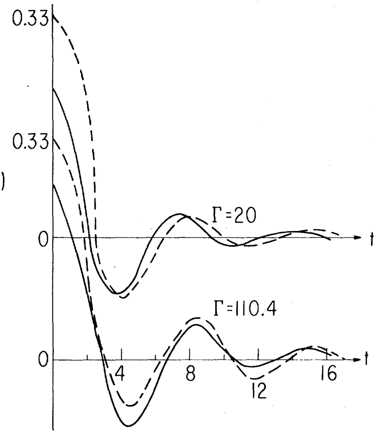

FIG. 1. Comparison of memory function obtained from molecular dynamics (dashed line) (Ref. 2) and from microscopic theory (solid line) for $\Gamma = 20$ and 110.4. The time scale is in units of $\omega_p{}^{-1}$ and $M(t)$ is in units of $\omega_p{}^{-2}$.

velocity autocorrelation function of a dense one-component classical plasma.

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[3] We adopt the standard notation with e the charge, n the number density, N the number of particles, T the temperature, k_B Boltzmann's constant, m the mass, and $\beta = 1/k_BT$. Γ is related to the "plasma parameter" ϵ by $\Gamma = \epsilon^{2/3}/3^{1/3}$, where $\epsilon = \beta e^2/D$ and $D = (4\pi\eta\beta e^2)^{-1/2}$.

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[10] See, for example, Ref. 5, Sect. VII.

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[12] We neglect $C_p - C_v$ compared to C_v. See Ref. 10.

Magnetic Resonance in Amorphous $TbFe_2$ and $GdFe_2$†

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We report measurements on the spin resonances in a-$TbFe_2$ and a-$GdFe_2$. A preliminary interpretation in terms of an oversimplified model involving clusters in a disordered material is also outlined.

Amorphous ferromagnetism has attracted considerable attention in recent years. For instance, a-$TbFe_2$ and a-$GdFe_2$ have been studied in great detail by Rhyne and co-workers using both neutron scattering[1] and static measurements.[2] Recently, Rhyne made available to us a few specimens of these materials. We have studied the spin resonances in a-$TbFe_2$ at several microwave frequencies between 9 and 31 GHz and at several temperatures between 77 and about 450 K. In a-$GdFe_2$ measurements have been made at several frequencies and $77 < T < 300$ K. The results are very anomalous and at present we can offer only rather rough explanations for some of the observations. The experiments were performed in conventional microwave spectrometers using setups described in earlier papers from this laboratory.[3] The samples were of millimeter size, chips or needles spark-cut from larger boules. However, one of the a-$TbFe_2$ samples was a sphere of diameter ~1 mm and one of the a-$GdFe_2$ specimens was a large parallelepiped ($5\times 3.5\times 1$ mm^3). The sample temperature was monitored by use of a copper-Constantan thermocouple affixed to the outside of the cavity. In a separate series of experiments using two thermocouples (one replacing the sample) we have checked that the sample temperature is within a degree of the thermocouple temperature.

Figure 1 is a typical recorder trace of the field derivative of the power absorption in a-$TbFe_2$ at 10.8 GHz and 300 K. Such a spectrum, i.e., one strong line (S_1) and three weak satellites (S_2 through S_4) on its high-field side, is essentially characteristic of all the samples studied. By repeated measurements the center of S_4 (i.e., H_4) was determined to about 100 Oe. H_4 varies roughly linearly with frequency (Fig. 2). However, *it is surprising that it is essentially independent of sample shape.* On the other hand the value of H_1 reflected the changes in the static demagnetizing field due to changes of size and shape. S_2 being very weak and lying rather close to S_1, its position (H_2) cannot be fixed without considerable analysis involving assumptions as to line shape, etc. However, in every case $H_2 > H_1$. At present, we have only rough measurements on H_3. Further, it should be noted that the intensity