Longitudinal and transversal collective modes in strongly correlated plasmas

P. Schmidt, G. Zwicknagel,* P. -G. Reinhard, and C. Toepffer
Institut für Theoretische Physik II, Universität-Erlangen, D-91058 Erlangen, Germany

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We investigate excitations in strongly coupled one-component plasmas with the help of molecular-dynamics computer simulations. As dynamical observables we study the dynamical structure factor, the velocity autocorrelation function, and the longitudinal and transversal current correlation functions. The results on these observables can be related to each other in the framework of a mode-coupling theory. The appearance of transversal collective shear modes indicates liquid behavior, and is a precursor for an eventual transition to a Wigner lattice. [S1063-651X(97)02312-X]

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The physics of a classical one-component plasma (OCP) with a neutralizing background is governed by a single parameter, the ratio Γ of the mean potential energy between the particles and their thermal energy $\beta^{-1}=k_BT$

$$\Gamma=(Ze)^2/(4\pi\varepsilon_0 a k_B T). \tag{1}$$

Here Ze is the charge of the particles, ε_0 the dielectric constant of the vacuum, k_B the Boltzmann constant,

$$a=(4\pi n/3)^{-1/3} \tag{2}$$

the Wigner-Seitz radius, and n the number density. Despite its simplicity the OCP already shows many of the static and dynamical properties of much more complicated many-body systems, in particular transitions from an ideal, gaseous, weakly coupled regime for $\Gamma \ll 1$ through a liquid regime [1] to the eventual Wigner crystallization [2] into a solid Coulomb lattice near $\Gamma=160$. In the intermediate liquid regime Hansen and co-workers performed molecular-dynamics (MD) computer simulations [3] and extracted the velocity autocorrelation function (VAF). Its power spectrum showed some features which could be readily explained, e.g., a diffusive regime dominated by single particle collisions for $\Gamma \lesssim 1$, and a collective peak near the plasma frequency at larger values of Γ. However, for $\Gamma \gtrsim 100$ another prominent peak appears at a low, nonzero frequency which could not be interpreted easily. Hansen, McDonald, and Pollock attempted to give a unified description of the velocity autocorrelation function in the liquid regime using the memory function formalism, but found this a very difficult task and did not pursue the matter further [4]. A somewhat more general approach in sort of a mode coupling model was investigated in Ref. [5]. This model dealt successfully with the impact of collective plasmon modes on the VAF. The problem to explain the additional broad low-energy peak at high Γ remains, however, open.

It is the aim of this paper to present a mode-coupling model which yields a satisfactory interpretation of the VAF in the strong-coupling regime. It can be shown that the low-frequency peak at strong coupling is associated with the occurrence of transversal acoustic excitations (or shear modes) in the liquid.

Before presenting the results of our MD simulations and discussing the mode-coupling model, we first want to introduce a few basic definitions, thereby using the conventional notations [6,7]. A key tool to analyze the dynamical properties of a plasma are the autocorrelation functions $\langle A(t)A^*(t')\rangle$ of a dynamical observable A. They can be calculated either as an average over time or as an ensemble average. We will consider here equilibrium autocorrelation functions of the following observables:

$$\text{single particle velocity } \ \vec{v}_1(t), \tag{3}$$

$$\text{single particle density } \ \rho_s(\vec{k},t)=e^{i\vec{k}\cdot\vec{r}_1(t)}, \tag{4}$$

$$\text{current operator } \ \vec{j}(\vec{k},t)=\sum_j \vec{v}_j(t)e^{i\vec{k}\cdot\vec{r}_j(t)}. \tag{5}$$

With the help of the current operator (5), the longitudinal

$$C_l(\vec{k},t):=N^{-1}\langle[\vec{k}\cdot\vec{j}(\vec{k},t)][\vec{k}\cdot\vec{j}(-\vec{k},0)]\rangle, \tag{6}$$

and transversal

$$C_t(\vec{k},t):=(2N)^{-1}\langle[\vec{k}\times\vec{j}(\vec{k},t)]\cdot[\vec{k}\times\vec{j}(-\vec{k},0)]\rangle, \tag{7}$$

current correlation functions can be constructed. In contrast to the functions (6) and (7), the VAF,

$$Z(t)=\langle\vec{v}_1(t)\vec{v}_1(0)\rangle/\langle\vec{v}_1(0)\vec{v}_1(0)\rangle \tag{8}$$

[where $\langle\vec{v}_1(0)\vec{v}_1(0)\rangle=3k_{\mathrm{B}}T/m$] involves the single-particle velocity (3) of a tagged particle rather than the coherent superpositions (5). Another quantity which is related to single-particle properties is the self-intermediate scattering function, which is the autocorrelation function of the single-particle density (4)

$$S_s(k,t)=\langle e^{i\vec{k}\cdot\vec{r}_1(t)}e^{-i\vec{k}\cdot\vec{r}_1(0)}\rangle. \tag{9}$$

Spectral properties are better seen after Fourier transformation into frequency space. This reads, e.g., for the VAF,

*Present address: Laboratoire de Physique des Gaz et des Plasmes, F-91405 Orsay, France.

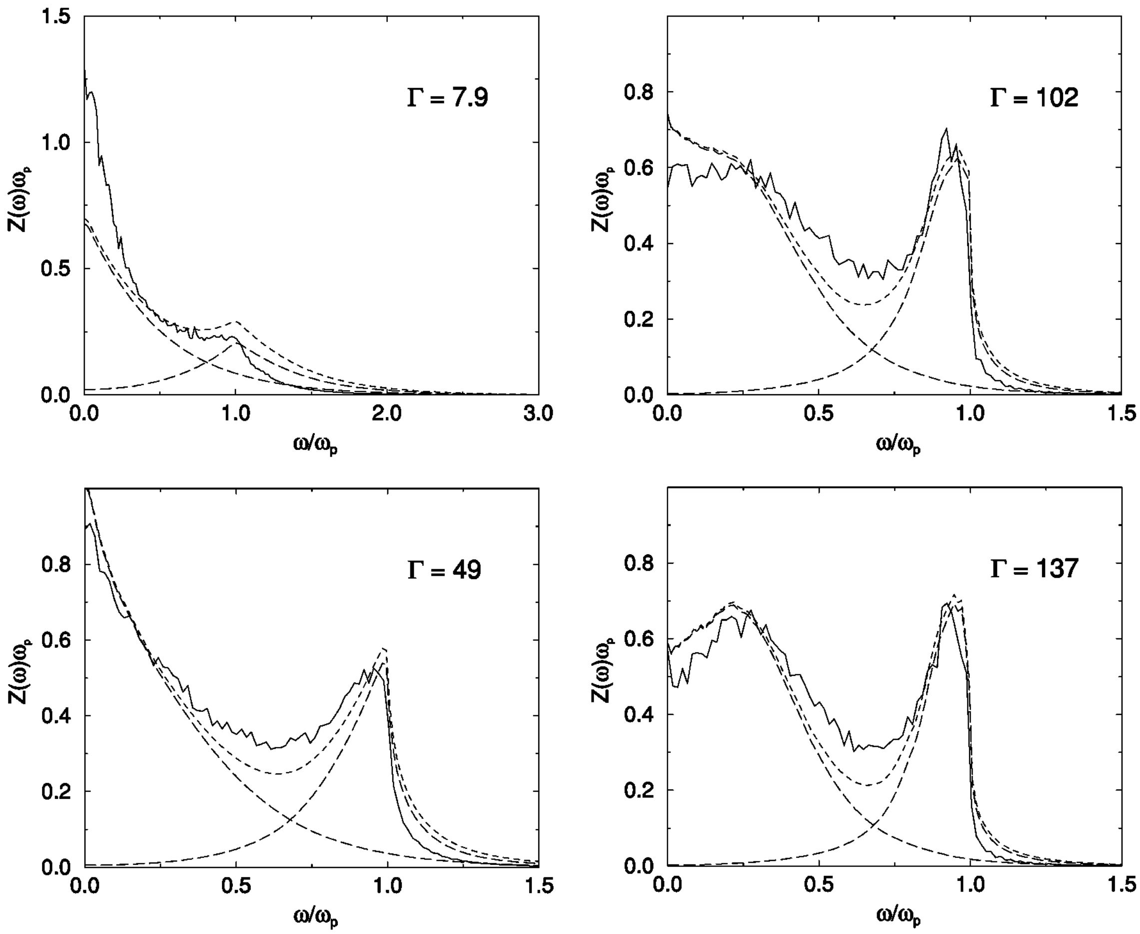

FIG. 1. MD results for the VAF $Z(\omega)$ (solid curve) in comparison to the mode-coupling model (16) for various values of the coupling. The curves with the long dashes show the separate contributions from C_l and C_t to Eq. (16) at high and low frequencies, respectively. Their sum is given by the curve with the short dashes. The MD simulations have been done with 500 particles in a cubic simulation box of length $L=12.8d$, where d is the interparticle distance.

$$Z(\omega)=(2\pi)^{-1}\int_{-\infty}^{\infty} Z(t)\exp(i\omega t)dt, \tag{10}$$

and similarly for all other correlation functions.

As mentioned in the Introduction, the properties of a plasma change with its coupling Γ, from a gaseous regime for weak coupling $\Gamma \ll 1$ over a liquid regime, up to a crystalline stage at very large coupling. These changes of regimes are reflected in the VAF, which shows signatures of a transition from a dominantly diffusive single-particle regime at small Γ to a collective regime at larger Γ. For $\Gamma \lesssim 5$, the VAF $Z(t)$ decays exponentially, except for very small times, and its Fourier transform $Z(\omega)$ has a purely diffusive peak at zero frequency. For increased coupling the decay of $Z(t)$ becomes oscillatory, and a peak near the plasmon frequency $\omega_p=(e^2n/m\varepsilon_0)^{1/2}$ starts to appear in $Z(\omega)$; see the MD results (full line) in the panel for $\Gamma=7.9$ in Fig. 1. The plasmon peak gains weight with further increasing Γ, and, at even larger Γ, a new broad peak at around 0.3 ω_p emerges, see the panels with $\Gamma>100$ in Fig. 1.

In an attempt to analyze their MD simulation results, Hansen and co-workers [4,6] showed that the first transition from a diffusive regime at $\Gamma \lesssim 5$ to the appearance of the plasmon peak in the VAF at larger Γ could be roughly interpreted in terms of a memory kernel formalism. The memory kernel model puts the division line between the diffusive and the oscillatory regime at around $\Gamma \sim 4$, in fair agreement with the empirical $\Gamma \sim 5$, as deduced from scanning the sequence of $Z(\omega)$. The memory kernel model is still too crude, however, to describe all details of the VAF emerging at strong coupling, because it fails to provide the coexistence of a diffusive peak and an increasingly competitive plasmon peak, as seen e.g., in panel $\Gamma=7.9$ of Fig. 1. Of course, any structure, even the appearance of the broad low-energy peak at large Γ, could be fitted in a memory kernel model when carrying further the continued fraction expansion to cover more poles. But such an expansion then becomes a merely mathematical tool, and the model gives little insight into physics beyond the VAF.

However, transversal collective modes have been studied in the quasilocalized charge formalism for the dielectric tensor [8]. Indeed, an inspection of the current-current autocorrelation functions (6) and (7) shows that the one-component plasma supports a transversal acoustic mode at $\Gamma \gtrsim 100$. As an example, in Fig. 2 we show the dispersion relations for the high-frequency plasmon mode and the low-frequency acoustic (shear) mode for a very strongly coupled system with $\Gamma=158$. It should be noted that the longitudinal and

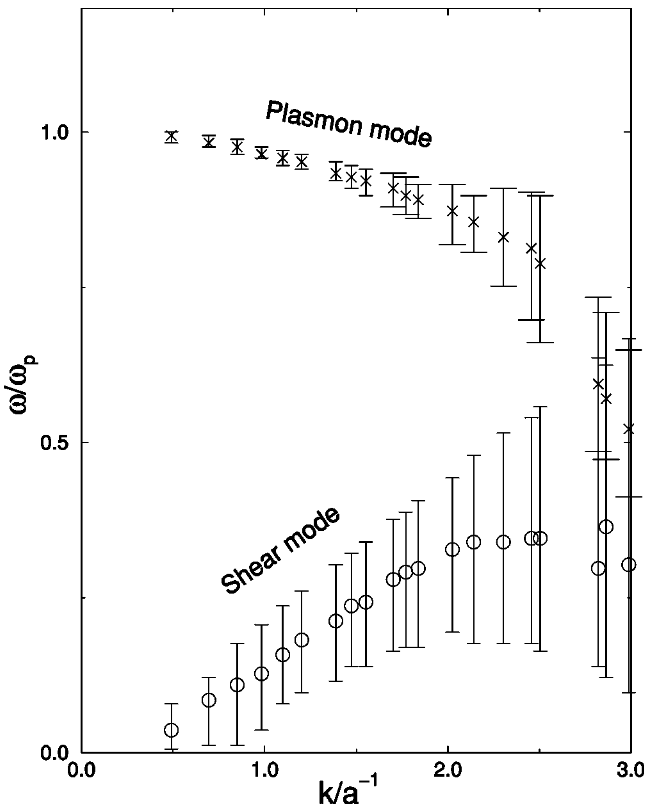

FIG. 2. Dispersion relations for the high-frequency plasma wave and the low-frequency acoustic mode obtained from a MD simulation at strong coupling, $\Gamma=158$. The current correlation functions $C_{l,t}(k,\omega)$ were smoothed using Savitzky-Golay filters [12]. As suggested in Ref. [7], a dispersion relation $\omega(k)$ is obtained from the maxima of $C_{l,t}(k,\omega)$ at fixed k. It is indicated by crosses for the plasmon mode, and by circles for the acoustic mode. The error bars mark the intervals where $C_{l,t}(k,\omega)$ is larger than 75% of the maximal value.

transversal modes are only reasonably well defined up to the first Brillouin zone $k \leqslant \pi/a$. Larger momenta correspond to distances below the interparticle spacing, where collective modes make little sense. But the two collective modes, acoustical and optical branch, are well developed at low k. It is to be expected that the acoustical branch has increasing impact on the VAF with increasing Γ, and it is obvious that one should relate that to the appearance of the broad low-energy peak in $Z(\omega)$ at the two larger Γ in Fig. 1. The feature of a broad peak could then be explained by the mixing of modes with different k and the linear growth of ω with k. In order to substantiate this conjecture, one needs a model which establishes a unique relation between these collective modes and the VAF, like the mode-coupling model [6,9]. This had already been discussed in connection with the VAF of strongly correlated plasmas in Ref. [5]; though without an acoustic mode and with a simplified input for the correlation functions. We will employ this mode-coupling model here, with proper emphasis on the acoustic modes, using fully consistent microscopic input for the necessary current-current correlation functions.

To introduce the mode-coupling model, we consider the time evolution of an observable A in a system with the Hamiltonian H which is described by $dA/dt = i\mathcal{L}A = -\{H,A\}$, where the Liouville operator $\mathcal{L}$ is used as abbreviation for the Poisson bracket $\{H,A\}$. Formal integration then yields $A(t)=\exp(it\mathcal{L})A(0)$. This full propagation, however, is rarely feasible and approximate treatments have to be invoked. One widely used method is the projection operator technique [10], where one chooses a projector $\mathcal{P}$ on the space of relevant observables, and replaces the exact propagator by

$$\exp(it\mathcal{L}) \to \mathcal{P}\exp(it\mathcal{L})\mathcal{P}. \tag{11}$$

In the following, we will employ the mode-coupling model of Ref. [6]. It is our aim to relate the VAF, Eq. (8), with the collective modes of the system. To that end one chooses, for the set of relevant operators,

$$B_{\alpha\vec{k}} = \rho_s(\vec{k})\, j^{\alpha}(-\vec{k}), \tag{12}$$

a combination of single-particle density ρ_s [see Eq. (4)] and collective current j^{α}, where $\alpha=x,y,z$ is the α component of the current; see Eq. (5). The projector is

$$\mathcal{P}\, A = \sum_{\alpha,\vec{k}} B_{\alpha\vec{k}}\, (B_{\alpha\vec{k}}, B_{\alpha\vec{k}})^{-1}\, (B_{\alpha\vec{k}}, A). \tag{13}$$

We will consider the particular case $A=v_1^{\gamma}$ (where $\gamma=x,y,z$) to compute the VAF. The scalar products can be calculated with the help of Eqs. (3), (4), and (5) as

$$(v_1^{\gamma}, B_{\alpha\vec{k}}) = \delta_{\gamma\alpha}\frac{k_B T}{m}, \tag{14}$$

$$(B_{\alpha\vec{k}}, B_{\alpha\vec{k}}) = N\frac{k_B T}{m}. \tag{15}$$

The VAF then becomes

$$Z(t) \simeq \frac{m}{3k_B T}\sum_{\gamma} (v_1^{\gamma}\mathcal{P}, \exp(it\mathcal{L})\mathcal{P}v_1^{\gamma})$$

$$= \frac{m}{3k_B T N^2}\sum_{\alpha}\sum_{\vec{k}} (B_{\alpha\vec{k}}, \exp(it\mathcal{L})B_{\alpha\vec{k}}).$$

The emerging correlation function of four variables is factorized according to

$$(B_{\alpha\vec{k}}, e^{it\mathcal{L}}B_{\alpha\vec{k}}) \simeq (\rho_s(\vec{k}), e^{it\mathcal{L}}\rho_s(\vec{k}))(j^{\alpha}(-\vec{k}), e^{it\mathcal{L}}j^{\alpha}(-\vec{k})),$$

i.e., the modes are assumed to propagate independently. This yields

$$\sum_{\alpha} (B_{\alpha\vec{k}}, e^{it\mathcal{L}}B_{\alpha\vec{k}}) = S_s(k,t)\big(C_l(k,t) + 2C_t(k,t)\big).$$

There then remain products of the self-intermediate scattering function and the current correlation functions. Changing the $\vec{k}$ summation into an integral, one finally obtains

$$Z(t) = \frac{m}{3k_{\mathrm{B}}T}\int \frac{d^3k}{8\pi^3} S_s(k,t)\,\frac{C_l(k,t)+2C_t(k,t)}{nk^2}. \tag{16}$$

The integral in Eq. (16) is divergent at large k. But it does not make sense to carry the collective currents that far. A cutoff $\propto 1/a$ is to be expected because the currents cannot resolve spatial details closer than the interparticle distance a. We tailor the cutoff to guarantee proper normalization of the VAF. Using $S_s(k,t=0)=1$ and $C_l(k,t=0)+2C_t(k,t=0)=(3k_BT/m)\ k^2$, we enforce $Z(t=0)=1$ by cutting the k integral at

$$k_c=(6\pi^2 n)^{1/3}=2.42/a, \tag{17}$$

i.e., by excluding modes with wavelength which are smaller than the mean distance between the particles.

Equation (16) together with the appropriate cutoff prescription is the essential result of the mode-coupling model. It establishes a relation between the collective currents C_l and C_t, together with the autocorrelation function of the single particle density S_s and the VAF $Z(\omega)$. It thus models the VAF as composed from the basic plasma modes, the longitudinal plasmon branch and the transversal acoustic branch. We now analyze the MD results for the VAF in terms of the mode-coupling model. To that end, we determine also the ingredients C_l, C_t, and S_s from the same MD simulations. In Fig. 1 we compare, at various coupling parameters Γ, the MD result for $Z(\omega)$ (solid line) with the results from the mode-coupling model. The separate contributions of C_l and C_t to $Z(\omega)$ [see Eq. (16)] are shown as curves with long dashes. The short dashes represent the mode-coupling model in total. For large coupling $\Gamma \gtrsim 100$ one can clearly distinguish the peak just below ω_p, resulting from the plasma mode and the low-frequency peak around $0.3\omega_p$, resulting from the acoustic mode. In this regime the sum of these contributions (short dashes in Fig. 1) agrees well with the solid curve. This demonstrates that the peak at low but finite frequency which arises for large Γ is due to the acoustic modes in the plasma. The agreement of the mode-coupling model with the MD data gets worse, however, for small coupling, as indicated for the case of $\Gamma=7.9$ in Fig. 1. And it fails completely for even smaller Γ. The technical difficulty is that the current correlation functions become broader in k space [4,11], which attaches too much weight near k_c to the integral in Eq. (16). The physical problem is that the mode-coupling model relies too much on the collective modes alone, and thus cannot properly take into account the single-particle processes which produce the dominant diffusive peak at small coupling.

To conclude: We have investigated the velocity autocorrelation function $Z(t)$ of a one-component plasma in the regime of strong coupling Γ. The results from MD simulations show first the well-known dominance of the plasmon oscillations over diffusive decay for strong coupling, with a transition point at around $\Gamma \sim 5$. A closer inspection of the Fourier transform $Z(\omega)$ at even larger $\Gamma \gtrsim 100$ shows, however, the appearance of a low-frequency peak in addition to the plasma peak and the (then nearly negligible) diffusive contribution. We have investigated the structure of the VAF at very large coupling with a mode-coupling model which establishes a relation between the VAF and the collective current-current correlation functions. In this way we could show that the broad low-frequency peak is due to the acoustic modes, which start to play a role in the liquidlike plasma at very large coupling. Taken together with earlier findings, this shows that three processes cooperate in the VAF with changing weights when changing the coupling: dominantly particle diffusion at small coupling, increasing importance of the plasmon oscillations for larger coupling with turning point at around $\Gamma \sim 5$, and additional appearance of the acoustic modes at $\Gamma \sim 100$.

There remains an open point in that the mode-coupling model cannot account for the broad diffusive background in $Z(\omega)$ at smaller coupling. It seems worthwhile to extend the mode-coupling model in a similar manner as has been done for fluids of uncharged particles.

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