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Dynamic form factor of two-component plasmas beyond the static local field approximation

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Abstract

The spectrum of ion density fluctuations in a strongly coupled plasma is described both within the static $G(k, 0)$ and high-frequency $G(k, \infty)$ local field approximation. By a direct comparison with molecular dynamics data, we find that for large coupling, $G(k, 0)$ is inadequate. Based on this result, we employ the Zwanzig–Mori memory function approach to model the Thomson scattering cross section, i.e. the electron dynamic form factor $S_{ee}(k, \omega)$ of a dense two-component plasma. We show the sensitivity of $S_{ee}(k, \omega)$ to three approximations for $G(k, \omega)$.

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(Some figures in this article are in colour only in the electronic version)

1. Introduction

We are interested in the dynamical properties of solid-density plasmas, which can be created with short-pulse lasers. Such plasmas may be strongly coupled, as characterized by a ratio of the potential to the kinetic energy per particle larger than one. The dynamical properties depend on the intricate details of various interaction processes that are affected by strong correlations. Furthermore, the dynamical properties can be used, in principle, as a diagnostic of the solid-density plasma if the scattered spectrum of radiation can be predicted. For example, in analogy with dilute-plasma diagnostic, Thomson scattering (TS) from free electrons may yield information about the density and temperature of the plasma [1] and, in turn, provide a delicate test of our understanding of dynamical and transport properties of strongly coupled systems. Recently, experiments have been performed in the strongly coupled plasma regime [2], where most theoretical approaches do not apply.

An important quantity is the spectrum of the electron density–density fluctuations, the so-called dynamic structure factor $S_{ee}(k, w)$, since it is directly probed in TS (here k and ω are, respectively, the wave vector and the frequency of the electron density fluctuations).

There have been a few descriptions of $S_{ee}(k, w)$ in the strongly coupled regime. For example, Boerker, Lee and Rogers (BLR) [3] have used a 'generalized Vlasov approximation' to predict the position of the collective modes. The applicability of this approach is investigated in section 2. By applying projection operator techniques to kinetic equations, Linnebur and Duderstadt (LD) [4] were able to generate exact equations describing density fluctuations in a two-component plasma in thermal equilibrium. The equations reduce to the coupled linearized Vlasov equations augmented by additional terms which characterize collisions; the different applications proposed so far [4, 5] also depend upon the various Coulomb logarithms, just as the more traditional collision models do. A similar approach was taken by Gregori *et al* [6] to describe the charge density fluctuations in several approximations. In both cases, however, correlations were accounted for in the Debye–Hückel approximation.

In this paper, we first illustrate the difficulty of constructing a theory of S_{ee} by studying the effect of coupling on the collective ionic dynamics in a simple model. We also introduce the concept of an effective interaction, as described through a local field correction (LFC). We show that the static LFC, as used by BLR, is not satisfactory for describing the effects of strong coupling on the TS cross section at moderate k and ω. The concept of LFC is intimately connected with the concept of memory functions that appears when the dynamics of physical quantities are written in terms of a generalized Langevin equation. In section 3, we use such a memory function approach, usually referred to as the Zwanzig–Mori (ZM) formalism [7, 8], to generate improved models of TS. The sensitivity of S_{ee} to the models is illustrated in a comparison of three different approximations. Note that here we ignore certain physical processes and corrections that may be important in some experiments such as atomic processes [9] or Compton scattering [10]. It is important to mention, however, that plasmas dense enough to be strongly coupled may be most easily diagnosed with short-wavelength radiation and the simplified model we employ here must be extended.

2. Local field corrections and ion-acoustic waves

In a two-component plasma, peaks occur in the electron–electron dynamic form factor $S_{ee}(k, \omega)$ as a result of the collective behaviour of electrons and ions. In particular, at low frequency, a peak occurs as a result of electrons following the collective motion of the ion subsystem to maintain quasineutrality. The underlying ion wave, the ion-acoustic wave (IAW), is likely to display the effects of strong coupling more prominently than the electron wave since the ions are often cooler and have higher charge states Z. It is therefore of interest to begin by studying the properties of this feature in the cross section, and we do so by considering the spectrum of ion density fluctuations $S(k, \omega)$.

To model this regime, we describe the plasma as a collection of particles, the screened ions, interacting through a Yukawa pair potential $v(r) = \frac{\Gamma}{\beta r}\exp(-\kappa r)$, where $\kappa^{-1} = (ak_{De})^{-1}$ is the electron screening length and $\Gamma = (Ze)^2/k_BTa$ is the Coulomb coupling parameter. In this section lengths are written in units of the ion-sphere radius $a = (3/4\pi n)^{1/3}$ (n is the particle density) and frequencies in units of the plasma frequency $\omega_i = \left(\frac{4\pi(Ze)^2 n}{m}\right)^{1/2}$. $S(k, w)$ can be evaluated using the fluctuation–dissipation theorem,

$$\omega_i S(k, w) = -\frac{2}{n\beta\omega}\,\mathrm{Im}\,\chi(k, w). \tag{1}$$

The response function $\chi(k, \omega)$ can be exactly written in terms of a reference system, $\chi^0(k, \omega)$, usually taken as the free particle system, and a frequency and wave vector dependent, complex effective potential $V(k, \omega) = v(k)(1 - G(k, \omega))$ such that

$$\chi(k,\omega) \equiv \frac{\chi^0(k,\omega)}{1 - v(k)(1 - G(k,\omega))\chi^0(k,\omega)}. \tag{2}$$

Here $G(k,\omega)$ is the so-called dynamic local field correction (LFC) and $v(k)$ is the Fourier transform of the pair potential and $\beta n v(k) = \frac{3\Gamma}{k^2+\kappa^2}$.

In the generalized Vlasov approximation, the effective potential $V(k,\omega)$ is taken to be real and ω-independent, which amounts to replacing the original pair interaction by an effective (renormalized) interaction, $V(k,\omega) = v_{\rm eff}(k)$. Within such an approximation, only static effects arising from strong coupling are taken into account. Although it may reproduce reasonably well the dispersion of collective modes, it is incapable of accounting correctly for the damping of these modes. In practice, $v_{\rm eff}(k)$ is chosen to obey specific sum rules (to the accuracy that the static properties, in particular the pair distribution, are known). Two common approximations are the static local field correction (SLFC) and high-frequency local field correction (HFLFC). In their approach, BLR used the SLFC, which amounts to replacing the pair interaction by $v_{\rm eff}(k) = v(k)(1 - G(k,0))$,

$$G(k,\omega) \to G(k,0) = 1 - \frac{1}{v(k)}\left(\frac{1}{S(k)} - 1\right). \tag{3}$$

This approximation guarantees that the sum rules $\langle\omega^0(k)\rangle$ and $\langle\omega^2(k)\rangle$ are exactly satisfied[1], where $\langle\omega^n(k)\rangle = \int \frac{{\rm d}\omega}{2\pi}\omega^n S(k,\omega)$.

Note that $\langle\omega^0(k)\rangle = S(k)$ is the static structure factor. The SLFC is directly related to the isothermal compressibility $\chi_T = \frac{\beta}{n}\lim_{k\to 0}\frac{1}{1+v(k)(1-G(k,0))}$ [11]. Thus, the LFC acts as a nonlocal (k-dependent) renormalization of the compressibility and, in turn, of the phase speed $c_L \propto \chi_T^{-1/2}$ of the longitudinal wave. The SLFC is thus expected to yield a very good estimate of the dispersion relation of the collective longitudinal mode as $k \to 0$. However, as we will see, its range of validity at higher k and the dependence of this range on the coupling parameter Γ is strongly limited. Indeed, as $k \to 0$, the dynamics of the modes are strongly affected both by processes involving time scales not short compared to the hydrodynamic characteristic times and by nonlocal effects, which arise when the range of correlations becomes of the order of k^{-1}.

In the HFLFC approximation, the strong coupling behaviour is treated in a high frequency approximation defined by

$$G(k,\omega) \to G(k,\infty) = 1 - 3\Gamma\frac{k^2+\kappa^2}{k^4}\left(\langle\omega^4\rangle(k)\big/\omega_i^4 - \frac{k^4}{3\Gamma^2}\right). \tag{4}$$

Now $\langle\omega^2(k)\rangle$ and $\langle\omega^4(k)\rangle$ are automatically satisfied, but the sum rule $\langle\omega^0(k)\rangle = S(k)$ is not. In particular, HFLFC does not yield the exact isothermal compressibility χ_T and, in turn, is surely incapable of describing the acoustic part of the dispersion relation near $k = 0$. However, as we will see, this simple argument does not make HFLFC a bad approximation of the dispersion relation of the IAW over the entire k-range.

In figure 1, we compare the dispersion relation of the longitudinal mode as obtained from molecular dynamics (MD) simulations [12] with calculations using the SLFC and HFLFC approximations. In both SLFC and HFLFC calculations, the dispersion relation has been obtained both from the peak of $S(k,\omega)$ (see equation (1)), and from the simple estimates

$$\omega(k)/\omega_i = k\sqrt{\frac{1 - G(k,0)}{k^2+\kappa^2}} \qquad \text{(SLFC)} \tag{5}$$

$$\omega(k)/\omega_i = k\sqrt{\frac{1 - G(k,\infty)}{k^2+\kappa^2}} \qquad \text{(HFLFC)}. \tag{6}$$

[1] The second-moment sum rule is independent of the potential. It is thus automatically satisfied, irrespective of the choice of the DLFC.

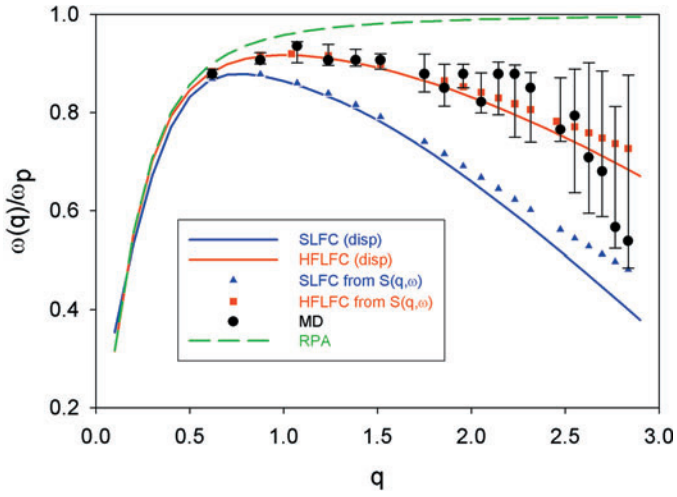

Figure 1. Dispersion relation for the longitudinal mode for a Yukawa system with $\Gamma = 144$ and $\kappa = 0.3$. Points are molecular dynamics results with error bars indicating the width at half the peak value. The curves correspond to the theoretical predictions: solid blue (red) is the SLFC (HFLFC) dispersion relation equation (5) (equation (6)); blue triangles (red) are the peaks of $S(k, \omega)$ in the SLFC (HFLFC) approximation. HFLFC leads to considerably better results than SLFC.

The latter expression, equation (6), can be shown to be closely related to mode dispersion in amorphous solids and liquids [13], and is sometimes referred to as the quasilocalized charge approximation (QLCA) [14]. In the example considered here the system is quite strongly coupled: $\Gamma = 144$, $\kappa = 0.3$, which corresponds to an effective (screened) coupling parameter $\Gamma_{\text{eff}} \equiv \Gamma \exp(-\kappa) \approx 106$ [12]. As compared with the MD results, one sees that HFLFC leads to considerably better results than SLFC, which was used in the BLR model. Note also that the dispersion relation is somewhat sensitive to using the peaks of $S(k, \omega)$ versus simple estimates. Even though HFLFC is better, it does not yield the correct compressibility relation for $k \to 0$. Only ω-dependent approximations for $G(k, \omega)$ can be expected to be consistent with the correct limits.

In this section we have considered the IAW spectrum within the Yukawa model only. In reality, plasmas are multicomponent systems for which the electron plasma wave also serves as a useful diagnostic. Moreover, the electron and ion dynamics are coupled and various collision processes affect them. We therefore need to generalize the ideas of this section to electron–ion systems. This is the goal of the next section.

3. Multicomponent theory

3.1. Theory

Here we consider a (neutral) dense strongly coupled TCP consisting of electrons e (charge $-e$, mass m_e, density n_e) and ions i ($Ze, m_i, n_i = n_e/Z$). For simplicity, the system is assumed to be in equilibrium (temperature $\beta = 1/k_B T$). Moreover, we treat the system classically, although the formalism presented here can be straightforwardly extended to the fully quantal case. In practice, of course, the TCP cannot be treated wholly classically since the ion–electron attraction leads to a collapse; the bare Coulomb potentials are usually replaced by effective potentials v_{ab} which account for diffraction and symmetry effects in an approximate way [15].

The ZM formalism [7, 8] gives the equations of motion for the fluctuations $\delta A_j(t)$ of predetermined relevant variables in the form of a generalized Langevin equation. This, in turn, leads to a memory function equation for the correlations between the fluctuations. When applied to the densities of particles $A_j = n_j$, this approach offers transparent schemes of approximation of the density fluctuation spectrum in a plasma. An attractive property of the memory function equation is its representation as a continued fraction [16, 17] whose terms depend uniquely on the moments of the correlations functions, the sum rules. The continued fraction representation permits one to make approximations, while satisfying several sum rules. Models for TS, i.e for $S_{ee}(k, \omega)$, are then deduced from the fluctuation–dissipation theorem.

The central quantity for TS is the electron–electron dynamic structure factor $S_{ee}(k, w)$. Within linear response theory, the fluctuation-dissipation theorem relates the dynamic structure factor to the electron–electron response function χ_{ee}

$$S_{ee}(k, w) = -\frac{2}{\rho\beta\omega} \operatorname{Im} \chi_{ee}(k, z = \omega) \tag{7}$$

where $\rho = n_e + n_i$ is the total number density of particles. In the following, we use the ZM formalism to generate models for χ_{ee}.

In view of the importance of the electron–ion coupling, it is worth writing the linear response of the TCP in terms of the response

$$\mathbf{A}(k, t) = \begin{pmatrix} \delta n_e(k, t) \\ \delta n_i(k, t) \end{pmatrix} \tag{8}$$

of vector of the Fourier components of the density fluctuations δn_e and δn_i. The information we require is contained in the correlation matrix $\tilde{a}(k, t)$ with elements $(a, b = e, i)$

$$\tilde{a}_{ab}(k, t) = \langle A_a(k, t), A_b(k, t = 0) \rangle \tag{9}$$

where $\langle \cdots \rangle$ denotes the statistical average. Indeed, according to linear response theory [11], if $\tilde{\chi}$ denotes the matrix of the density–density response functions (χ_{ab}), then the Laplace transforms $\tilde{\chi}(k, \omega)$ and $\tilde{a}(k, z)$ are related such that (the dot denotes the matrix product and $\tilde{1}$ the unit matrix)

$$\tilde{\chi}(k, z) \cdot \tilde{\chi}^{-1}(k) = \mathrm{i}z\tilde{a}(k, z) + \tilde{1}. \tag{10}$$

As stated before, the ZM formalism allows one to write the equation of motion of the correlation matrix $\tilde{a}(k, t)$ which, in the Laplace space, can be represented in the form of a continued fraction (here of matrices) such as [18]

$$\begin{aligned} \tilde{a}(k, z) &= (-\mathrm{i}z\tilde{1} + \tilde{b}_1(k, z) \cdot \tilde{\Delta}_1(k))^{-1} \\ \tilde{b}_1(k, z) &= (-\mathrm{i}z\tilde{1} + \tilde{b}_2(k, z) \cdot \tilde{\Delta}_2(k))^{-1}. \\ &\cdots \end{aligned} \tag{11}$$

In equation (11), the coefficient matrices $\tilde{\Delta}_\nu(k)$ depend only on the static properties of the system through the sum rules

$$\left\langle \omega_{ab}^n(k) \right\rangle = \int \frac{\mathrm{d}\omega}{2\pi} \omega^n S_{ab}(k, \omega). \tag{12}$$

In particular, in matrix notation,

$$\tilde{\Delta}_1(k) = \langle \omega^2(k) \rangle \cdot \langle \omega^0(k) \rangle^{-1} \tag{13}$$

$$\tilde{\Delta}_2(k) = \langle \omega^4(k) \rangle \cdot \langle \omega^2(k) \rangle^{-1} - \langle \omega^2(k) \rangle \cdot \langle \omega^0(k) \rangle^{-1}. \tag{14}$$

Thus, the determination of $\tilde{a}(k, z)$ is transformed to that of the matrices $\tilde{\Delta}_\nu$.

This representation of the correlation matrix provides a suitable starting point for developing approximations of $\tilde{\chi}$. This fact was exploited by Hong and Kim (HK) for a one-component plasma [18]. First, by generalizing the concept of LFC discussed in the previous section, it is convenient to express the matrix $\tilde{\chi}$ in terms of a matrix $\tilde{V}^{\rm eff}(k,z)$ such that

$$\tilde{\chi} = \tilde{\chi}_0 \cdot (1 - \tilde{V}^{\rm eff} \cdot \tilde{\chi}_0)^{-1} \tag{15}$$

where $\tilde{\chi}_0$ corresponds to the response function matrix for a free-particle TCP. Applying the continued fraction equation (11), one obtains

$$\begin{aligned}\tilde{V}^{\rm eff}(k,z) &= \tilde{\chi}_0^{-1}(k,z) - \tilde{\chi}^{-1}(k,z) \\ &= \left(\tilde{\chi}_0^{-1}(k) - \tilde{\chi}^{-1}(k)\right) + iz\tilde{\chi}_0^{-1}(k) \cdot \tilde{\Delta}_{0,1}^{-1} \cdot (\tilde{b}_2 \cdot \tilde{\Delta}_2 - \tilde{b}_{0,2} \cdot \tilde{\Delta}_{0,2})\end{aligned} \tag{16}$$

for the matrix $\tilde{V}^{\rm eff}$. The effects of the strong electron–ion coupling on the TS cross section appears through the effective potential as ($z = \omega$)

$$S_{ee}(k,\omega) = -\frac{2}{n\beta\omega}\,{\rm Im}\left\{\frac{\chi_{0,e}(k,\omega)\left[1 - V_{ii}^{\rm eff}(k,\omega)\chi_{0,i}(k,\omega)\right]}{D(k,\omega)}\right\}$$

where

$$\begin{aligned}D(k,\omega) &= \left[1 - V_{ee}^{\rm eff}(k,\omega)\chi_{0,e}(k,\omega)\right]\left[1 - V_{ii}^{\rm eff}(k,\omega)\chi_{0,i}(k,\omega)\right] \\ &\quad - \left(V_{ei}^{\rm eff}(k,\omega)\right)^2 \chi_{0,e}(k,\omega)\chi_{0,i}(k,\omega).\end{aligned} \tag{17}$$

3.2. Approximations for $V^{\rm eff}(k,\omega)$

In this section, we describe four parameter-independent approximations which can be devised from equation (16).

- *RPA with semiclassical potentials.* If we choose

  $$V_{ab}^{\rm eff}(k,w) = v_{ab}(k) \tag{18}$$

  we are led to the Vlasov approximation. Effective semiclassical potentials v_{ab} can be used instead of the bare Coulomb potentials [7].
- *Static local field correction (SFLC).* If we choose to satisfy $\langle\omega_{ab}^0(k)\rangle$ and $\langle\omega_{ab}^2(k)\rangle$ (equation (12)) we are led to

  $$\tilde{V}^{\rm eff}(k,w) = \tilde{V}^{\rm eff}(k,0) = \left(\tilde{\chi}_0^{-1}(k) - \tilde{\chi}^{-1}(k)\right). \tag{19}$$

  The approximation, which is the generalization of equation (3), amounts to setting $\tilde{b}_2 \cdot \tilde{\Delta}_2 = \tilde{b}_{0,2} \cdot \tilde{\Delta}_{0,2}$ in equation (16). The matrix $\tilde{\chi}(k) = \tilde{\chi}(k, \omega = 0)$ is given in terms of the static form factor $S_{ab}(q)$ as

  $$\chi_{ab}(k) = -\beta\rho S_{ab}(q). \tag{20}$$

  In practice, the $S_{ab}(q)$ can be calculated by using the multicomponent hypernetted chain equations.
- *High-frequency local field correction (HFLFC).* If we choose to satisfy $\langle\omega_{ab}^2(k)\rangle$ and $\langle\omega_{ab}^4(k)\rangle$ we are led to

  $$V_{ab}^{\rm eff}(k,\omega) = V_{ab}^{\rm eff}(k,\infty). \tag{21}$$

  As in equation (4), the effective potential can be expressed uniquely in terms of the second- ($n = 2$) and fourth-order ($n = 4$) sum rules [19]. For strong coupling we anticipate that (21) is superior to (19), as illustrated in figure 1.

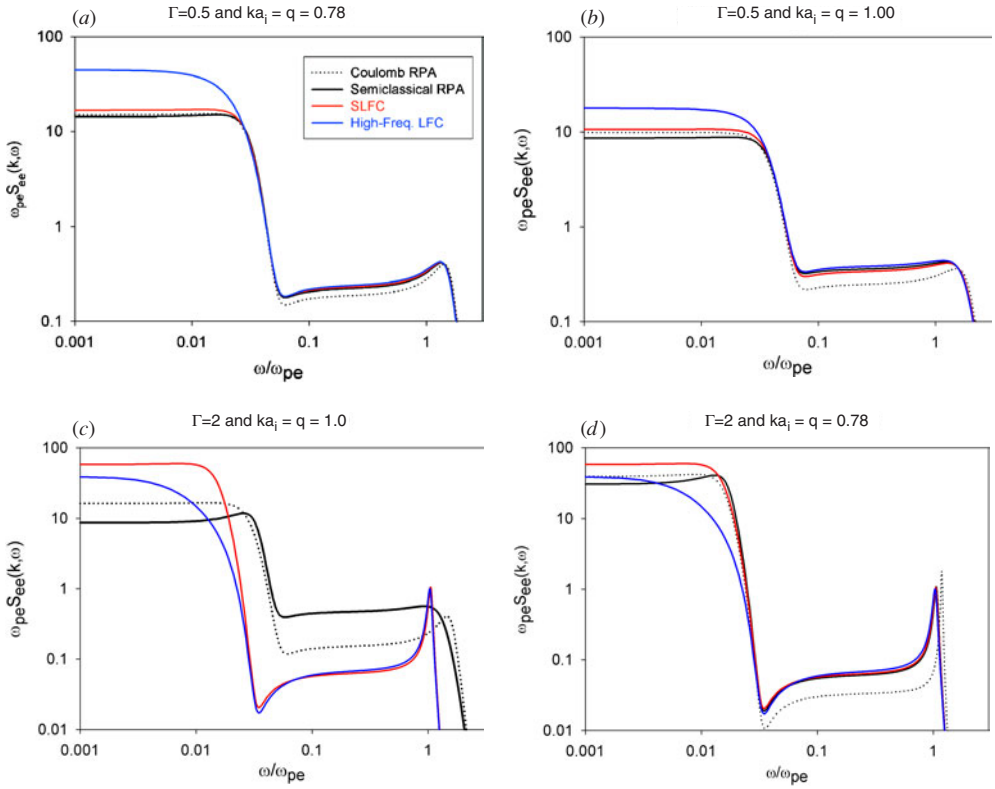

Figure 2. Electron dynamic form factor $S_{ee}(k, \omega)$ plotted against frequency normalized to ω_{pe} (double logarithmic scale) of hydrogen with $\Gamma = 0.5$ (upper part) and $\Gamma = 2$ (lower part).

A further step could be achieved by imposing the three sum rules $\langle \omega^0_{ab}(k) \rangle$, $\langle \omega^2_{ab}(k) \rangle$ and $\langle \omega^4_{ab}(k) \rangle$ to be satisfied. To this end, one could follow for instance the recipe given by HK in [18] and extend it to encompass the TCP. This approximation amounts to setting $\tilde{b}_2 = \tilde{b}_{f,2}$, where $\tilde{b}_{f,2}$ can be calculated from $\tilde{\chi}_0(k, z)$ using equations (10) and (11). Note that recently Wierling *et al* [20] have developed the HK approach for OCPs in terms of an unknown damping parameter; results of their fits to MD data indicate that there is no obvious trend in the damping parameter with variations in coupling. Another approach to satisfy the three sum rules has been given by Ichimaru *et al* [21]; unfortunately, this introduces three unknown functions $\tau_i(k)$. The approximations SLFC and HFLFC are the limits $\tau_i(k) \to 0$ and $\tau_i(k) \to \infty$, respectively.

As an illustration of the sensitivity of the results on the different approximations, we compare in figure 2 the dynamic structure factor $S_{ee}(k, \omega)$ for hydrogen as obtained from calculations using the four approximations: RPA with semiclassical potentials [15], SLFC, HFLFC and HK. Two Coulomb coupling parameters $\Gamma = 0.5$ and $\Gamma = 2$ and, for each Γ, two wave vectors $k = 0.78/a_i$ and $k = 1/a_i$ are considered. The above models require the first sum rules $\langle \omega^n_{ab}(k) \rangle$ as input information ($n = 0, 2$ for SLFC, $n = 2, 4$ for HFLFC and $n = 0, 2, 4$ for HK). This is accomplished here from the solution of the multicomponent hypernetted chain (HNC) equations [11]. Although the differences in the approximations are not large for $\Gamma = 0.5$, they are significant for $\Gamma = 2$ and mostly for the ion-acoustic feature.

4. Conclusion

The importance and range of applicability of TS as a plasma diagnostic depend on the accuracy of the theory. The difficulty of constructing a theory of TS for dense strongly coupled plasmas has been illustrated in a simple model of the IAW. We have shown that the static local field correction, as used by BLR, needs improvement in order to describe the effects of the strong coupling. To this end, we have compared the dispersion relation obtained with molecular dynamics simulations to results given by both the static and high-frequency local field approximations. In particular, for the case studied, HFLFC compares far better to MD results than SLFC. An open question is the validity of either SLFC or HFLFC for moderate coupling at finite k, ω.

Then, we have found it suitable to base the development of a theory of TS in strongly coupled TCP on the ZM formalism. In this way, one can easily satisfy the continuity equation, the compressibility sum rule and the fourth frequency moment, irrespective of the choice of the detailed form of the function $b_2(k)$ in equation (16).

The particular approximations studied were presented to illustrate the potential of the ZM approach, more particularly of the continued fraction representation, for modelling the TS cross section. More refined models will be developed in a future publication [19] which will allow for arbitrary ion charge Q and for two-temperature plasmas ($T_i \neq T_e$). Direct comparison with MD data will allow us to assess various models for the diagnostics of dense strongly coupled plasma.

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