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Dynamical mean-field theory for Coulomb systems: A novel approach

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We develop a novel approach to dynamical particle correlations in a classical one-component plasma system. A self-consistent integral equation is obtained for the dynamical local field to be determined. We show that this dynamical local field possesses the following properties: (i) it simultaneously satisfies the conservation and compressibility sum rules; (ii) it has the requisite constant values depending on frequency for large wave numbers; and (iii) it develops an algebraic tail in the high-frequency limit for small wave numbers.

We have been witnessing surging activity in the study of strongly coupled Coulomb systems.[1] A simple model of such systems, where the effect of strong correlations may be studied in a detailed manner, is the classical one-component plasma (OCP) system, where charged particles interact with each other in a neutralizing background. The OCP system is of interest in its own right as a representation of ions dispersed in a background of a neutralizing sea of degenerate electrons, or, even though classical, as a paradigm for the behavior of a degenerate electron gas at metallic densities. The strength of the interaction can be characterized by the plasma parameter $\gamma=e^2\kappa/kT=\kappa^3/4\pi n$, the inverse of the electron number in the Debye sphere, or by $\Gamma=e^2/akT$ [$\kappa=(4\pi e^2 n/kT)^{1/2}$ is the Debye wave number, and a is the interparticle distance or Wigner-Seitz radius.] In the weak-coupling limit γ and in the strong-coupling limit Γ can be identified as the ratio of the average potential energy to the average kinetic energy of the particles. Replacement of kT by $\frac{2}{5}\varepsilon_F$ (Fermi energy) in the case of the zero-temperature degenerate electron gas provides the correspondence between the conventionally used $r_s(=a/a_{\mathrm{Bohr}})$ and Γ by $\Gamma\rightarrow 1.36r_s$. [A different scaling results from establishing a correspondence between the r_s and Γ values where the crystallization of the unpolarized electron gas ($r_s^m\cong 80$) and the crystallization of the classical OCP ($\Gamma^m=178$) occurs: this yields $\Gamma\rightarrow 2.23r_s$.]

Central to the description of the properties of a correlated Coulomb system is the frequency and wave-number-dependent dielectric response function $\varepsilon(\mathbf{k}\omega)$. Various nonperturbative approximation schemes have been proposed for the calculation of $\varepsilon(\mathbf{k}\omega)$ both in the case of the electron gas[2–4] and of the classical OCP.[5,6] All these approximations can be classified in terms of their treatment of the so-called local-field factor $G(\mathbf{k}\omega)$ (to be discussed below) which, in turn, defines an "effective potential" $\phi(k)[1-G(\mathbf{k}\omega)]$ [$\phi(k)=4\pi e^2/k^2$ is the bare Coulomb potential]. For static properties particle correlations seem to be described quite accurately by a dielectric function with a static local field $G(\mathbf{k}\omega)=G(\mathbf{k})$. Yet, at the dynamic level, they are less satisfactorily explained through this kind of approximation. The failure can be remedied by restoring the dynamical nature of the local field. In this article, we present an alternative scheme for determining the dynamical local field self-consistently. This scheme is based on the recognition of the importance of the quadratic response function in the kinetic description of the system.[5] Although, in this respect, the philosophy of the present work parallels that of Ref. 5, it deviates fundamentally from it in its architecture. Reference 5 uses the velocity average approximation to relate the quadratic response function to the linear one: Here this is accomplished through the moment equation of motion method. Reference 5 approximates the quadratic response function through the "dynamical superposition approximation": here only a "reducible" structure [Eq. (7b)] is assumed for the quadratic response function. More importantly, the alternative approach seems to be able to provide results and meet consistency requirements in areas where the earlier approximation[5] was lacking.

Our starting point is the first equation of the Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) hierarchy for the distribution function $F(\mathbf{v};\mathbf{r}t)$ integrated over velocities[7] and differentiated with respect to time. The resulting equation for the density ρ is

$$\frac{\partial^2}{\partial t^2}\langle\rho(\mathbf{r}t)\rangle^{(1)}-\int d^3v\left[\mathbf{v}\cdot\frac{\partial}{\partial\mathbf{r}}\right]^2F^{(1)}(\mathbf{v};\mathbf{r}t)=\frac{n}{m}\frac{\partial^2\hat{\Phi}(\mathbf{r}t)}{\partial r^2}+\frac{1}{m}\frac{\partial}{\partial\mathbf{r}}\int d^3r'\frac{\partial}{\partial\mathbf{r}'}\phi(\mathbf{r}-\mathbf{r}')\langle\rho(\mathbf{r}t)\rho(\mathbf{r}'t)\rangle^{(1)} \tag{1}$$

valid to the first order of the external field $\hat{\Phi}$. (The superscripts refer to first-order perturbed quantities; $\rho^{(1)}$ is the first-order density response of the system, $\langle\rho\rho\rangle^{(1)}$ is the first-order two-point density-density correlation response.) After Fourier transformation, Eq. (1) becomes (ω_p is the plasma frequency)

$$(\omega^2-\omega_p^2)\chi(\mathbf{k}\omega)=\frac{nk^2}{m}+\lambda(\mathbf{k}\omega)+\sum_{\mathbf{q}\,(\neq 0)}\phi(\mathbf{q})\frac{\mathbf{k}\cdot\mathbf{q}}{m}\Xi(\mathbf{k}-\mathbf{q},\mathbf{q},\omega)\,, \tag{2}$$

where the linear density, linear kinetic, and double-density response functions are defined as[5]

$$\chi(\mathbf{k}\omega)\hat{\Phi}(\mathbf{k}\omega)=\rho^{(1)}(\mathbf{k}\omega)\ ,$$
$$\lambda(\mathbf{k}\omega)\hat{\Phi}(\mathbf{k}\omega)=\int(\mathbf{k}\cdot\mathbf{v})^2F^{(1)}(\mathbf{v};\mathbf{k}\omega)d^3v\ , \tag{3}$$
$$\Xi(\mathbf{k}-\mathbf{q},\mathbf{q},\omega)\hat{\Phi}(\mathbf{k}\omega)=\langle\rho_{\mathbf{k}-\mathbf{q}}\rho_{\mathbf{q}}\rangle^{(1)}(\omega)\ .$$

The double-density response function[8] Ξ may be replaced by the quadratic-density response function $\chi(\mathbf{k}_1\omega_1;\mathbf{k}_2\omega_2)$ through the quadratic fluctuation dissipation theorem[5,9] (FDT)

$$\Xi(\mathbf{k}-\mathbf{q},\mathbf{q},\omega)=\frac{-2}{\beta}\int d\mu\,\delta_-(\mu)[\chi(\mathbf{k}-\mathbf{q}\mu;\mathbf{q}\omega-\mu)+\chi(\mathbf{k}-\mathbf{q}\omega-\mu;\mathbf{q}\mu)] \tag{4}$$

in the classical limit.

In the random-phase approximation (RPA), which is the simplest mean-field approximation, the linear density, kinetic, and quadratic response functions have the structure

$$\chi_{\text{RPA}}(\mathbf{k}\omega)=\frac{\chi_0(\mathbf{k}\omega)}{\epsilon(\mathbf{k}\omega)}\ , \tag{5a}$$

$$\lambda_{\text{RPA}}(\mathbf{k}\omega)=\frac{\lambda_0(\mathbf{k}\omega)}{\epsilon(\mathbf{k}\omega)}\ , \tag{5b}$$

$$\chi_{\text{RPA}}(\mathbf{k}_1\omega_1;\mathbf{k}_2\omega_2)=\frac{\chi_0(\mathbf{k}_1\omega_1;\mathbf{k}_2\omega_2)}{\epsilon(\mathbf{k}_1\omega_1)\epsilon(\mathbf{k}_2\omega_2)\epsilon(\mathbf{k}\omega)}\ ,\qquad \mathbf{k}=\mathbf{k}_1+\mathbf{k}_2,\quad \omega=\omega_1+\omega_2\ , \tag{5c}$$

where

$$\chi_0(\mathbf{k}\omega)=\beta\int d^3v\frac{\mathbf{k}\cdot\mathbf{v}F(v)}{\omega-\mathbf{k}\cdot\mathbf{v}+io}\ , \tag{6a}$$

$$\lambda_0(\mathbf{k}\omega)=\beta\int d^3v\frac{(\mathbf{k}\cdot\mathbf{v})^3F(v)}{\omega-\mathbf{k}\cdot\mathbf{v}+io}=-nk^2/m+\omega^2\chi_0(\mathbf{k}\omega)\ , \tag{6b}$$

$$\chi_0(\mathbf{k}_1\omega_1;\mathbf{k}_2\omega_2)=\frac{\beta}{2m}\int d^3v\frac{F(v)}{(\omega-\mathbf{k}\cdot\mathbf{v}+io)^2}\times\left[\frac{(\mathbf{k}\cdot\mathbf{k}_1\mathbf{k}_2\cdot\mathbf{v})}{\omega_2-\mathbf{k}_2\cdot\mathbf{v}+io}+\frac{(\mathbf{k}\cdot\mathbf{k}_2\mathbf{k}_1\cdot\mathbf{v})}{\omega_1-\mathbf{k}_1\cdot\mathbf{v}+io}\right]\ , \tag{6c}$$

and

$$\epsilon(\mathbf{k}\omega)=1-\phi(\mathbf{k})\chi_0(\mathbf{k}\omega)\ . \tag{6d}$$

In the various more sophisticated mean-field approximations,[2-4] one assumes that the particle correlations still can be described by a single effective mean field and the particles move freely in such a mean field. Thus the structure of the linear-response functions is the same as that given by Eq. (5), except this time the screening is provided by the screening function $\Delta(\mathbf{k}\omega)$ augmented by the dynamical local field G,

$$\Delta(\mathbf{k}\omega)=1-\phi(\mathbf{k})[1-G(\mathbf{k}\omega)]\chi_0(\mathbf{k}\omega)\ , \tag{7a}$$

rather than by the dielectric function $\epsilon(\mathbf{k}\omega)$. In a similar vein, the quadratic response function can be assumed[10,11] to have the structure (5c) with appropriate substitution $\epsilon(\mathbf{k}\omega)\rightarrow\Omega(\mathbf{k}\omega)$

$$\chi(\mathbf{k}_1\omega_1;\mathbf{k}_2\omega_2)=\frac{\chi_0(\mathbf{k}_1\omega_1;\mathbf{k}_2\omega_2)}{\Delta(\mathbf{k}_1\omega_1)\Delta(\mathbf{k}_2\omega_2)\Delta(\mathbf{k}\omega)}\ . \tag{7b}$$

This latter observation combined with Eq. (4) provides the basis for the present approximation.

Substituting the modified Eq. (5) and Eq. (4) into Eq. (2), we obtain the relationship for the dynamical local field:

$$G(\mathbf{k}\omega)\chi_0(\mathbf{k}\omega)=\frac{1}{N}\sum_{\mathbf{q}\,(\neq 0)}\frac{\mathbf{k}\cdot\mathbf{q}}{q^2}\frac{2}{\beta}\int d\mu\,\delta_-(\mu)\left[\frac{\chi_0(\mathbf{k}-\mathbf{q}\mu;\mathbf{q}\omega-\mu)}{\Delta(\mathbf{k}-\mathbf{q}\mu)\Delta(\mathbf{q}\omega-\mu)}+\frac{\chi_0(\mathbf{k}-\mathbf{q}\omega-\mu;\mathbf{q}\mu)}{\Delta(\mathbf{k}-\mathbf{q}\omega-\mu)\Delta(\mathbf{q}\mu)}\right]\ . \tag{8}$$

Equations (8) and (7) constitute a self-consistent integral equation for the determination of dynamical local field. This integral equation is four-dimensional and highly nonlinear so that its solution is certainly not an easy matter and has to be sought numerically. However, with some further assumptions, it can be simplified considerably. By using the explicit representation (6c) of χ_0 in Eq. (8), the integration over the frequency can be carried out[10] with the aid of the Cauchy contour technique. The resulting expression contains contributions both from individual particle excitations (originating from χ_0) and from plasmon excitations (originating from the zeros of the Δ denominators). We argue[10] that these latter are negligible both for $\omega\rightarrow 0$ and $\omega\rightarrow\infty$ (as is obvious from the satisfaction of the low-frequency and high-frequency sum rules, as discussed below). For intermediate frequencies, especially in the vicinity of the plasma frequency, the plasmon pole contribution (physically corresponding to mode-mode interaction) may not be insignificant; it can, however, always be added to the expression derived below, without affecting its principal features. With this qualification, we obtain a relatively simple expression for the dynamical local field:

$$G(\mathbf{k}\omega)=-\frac{1}{N}\sum_{\mathbf{q}\,(\neq 0)}\frac{(\mathbf{k}\cdot\mathbf{q})^2}{k^2q^2}\left[\frac{1}{\Delta(\mathbf{k}-\mathbf{q}0)\Delta(\mathbf{q}\omega)}-\frac{1}{\Delta(\mathbf{k}-\mathbf{q}\omega)\Delta(\mathbf{q}0)}\right]-\frac{1}{N}\sum_{\mathbf{q}\,(\neq 0)}\frac{\mathbf{k}\cdot\mathbf{q}}{q^2}\frac{1}{\Delta(\mathbf{q}0)\Delta(\mathbf{k}-\mathbf{q}\omega)}\ . \tag{9}$$

This expression has a number of interesting features that we now proceed to list.

(i) In the static ($\omega=0$) limit, the application of the FDT shows that $1/\Delta(\mathbf{k}0)=S(\mathbf{k})$, the static form factor. Then, in this limit, Eq. (9) can be expressed as

$$G(\mathbf{k}0)=-\frac{1}{N}\sum_{\mathbf{q}\,(\neq 0)}\frac{\mathbf{k}\cdot\mathbf{q}}{q^2}S(\mathbf{k}-\mathbf{q})S(\mathbf{q})\ , \tag{10}$$

which can be identified as the Totsuji-Ichimaru (TI) expression[12] for the static mean field. Since the TI theory is known to provide a reasonably good description for the OCP, this is a satisfactory result. Moreover, the TI theory satisfies the compressibility sum rule for low coupling, which then ensures that the present theory enjoys the same property.

(ii) For high frequencies, Eq. (9) reduces to

$$G(\mathbf{k}\infty)=-\frac{1}{N}\sum_{\mathbf{q}\,(\neq 0)}\frac{(\mathbf{k}\cdot\mathbf{q})^2}{k^2q^2}[S(\mathbf{k}-\mathbf{q})-S(\mathbf{q})]\ , \tag{11}$$

as required by the high-frequency ω^{-4} (third frequency moment) sum rule.[13] To the best of our knowledge, this is the first time a dynamical theory has been able to satisfy the leading sum rules both at high and low frequencies.

(iii) At large wave number, carrying out the integration over the solid angle, we may show that the dynamical local field is given by

$$\begin{aligned}\lim_{k\to\infty} G(\mathbf{k}\omega)&=-\frac{2}{3N}\sum_{\mathbf{q}\,(\neq 0)}\left[\frac{1}{\Delta(\mathbf{q}0)}-1\right]\\ &\quad-\frac{1}{3N}\sum_{\mathbf{q}\,(\neq 0)}\left[\frac{1}{\Delta(\mathbf{q}\omega)}-1\right]\\ &=\begin{cases}\frac{2}{3}[1-g(0)], & \omega\to\infty\\ 1-g(0), & \omega\to 0\end{cases}\ ,\end{aligned} \tag{12}$$

where $g(0)\equiv g(r=0)$ represents the probability of finding two electrons at the same place. These results are in agreement with exact requirements derived by Niklasson and Shaw in the appropriate limits.[14]

(iv) The iterative solution of the integral equation based on the RPA input $G=0$ leads to the first iteration which for $k\to 0$ exhibits the algebraic tail

$$\begin{aligned}\mathrm{Im}[G(\mathbf{k}\omega)-G(\mathbf{k}\infty)]&=\frac{2\gamma k^2}{5\omega\pi}\left[\frac{\pi}{2}\right]^{1/2}\int dx\, xe^{-x^2/2}\\ &=\frac{\gamma k^2}{5\omega}\left[\frac{2}{\pi}\right]^{1/2}\end{aligned} \tag{13}$$

in the high-frequency limit. We may show that this algebraic tail prevails to any order of iteration and indeed is a feature of the self-consistent solution of Eq. (9). We note that the presence of the algebraic tail in ImG leads to an algebraic tail (k^4/ω^5) in the imaginary part of the density response function, in agreement with the previously obtained results of the perturbation calculations.[15] The immediate consequence of such a behavior is that any frequency moment higher than the third is divergent; this is corroborated again by the appearance of anomalous powers ($|\omega|^{-5}$) in the high-frequency expansion of the perturbative expression.[15,16]

(v) The relationship (9) ensures that the plus-function characters both of $G(\omega)$ and $\Delta^{-1}(\omega)$ are consistently maintained.

The integral equation (9) possesses the unique feature that in view of Eq. (10) it allows an iterative procedure, based on the *exact static* solution (obtained from HNC or Monte Carlo calculations) and leading to a frequency-dependent dynamical $G(\mathbf{k}\omega)$. This avenue for the actual calculation of $G(\mathbf{k}\omega)$ seems to be especially suitable for strong coupling.

In summary, we have developed a new approach to the dynamical correlations in Coulomb systems. An integral equation is established for the dynamical local field to be determined self-consistently. We find that our dynamical local field can simultaneously satisfy the conservation and compressibility sum rules. For large wave numbers, we show that the dynamical local field approaches the requisite constant values depending on frequency. For small wave numbers, we further show that the dynamical local field possesses an algebraic tail in the high-frequency limit.

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