

New Approach to a Strongly Coupled Electron Plasma

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A new approach to strongly coupled plasmas is proposed by a scheme linking the linear and quadratic polarizabilities through a self-consistency requirement. The ingredients of the theory are (a) replacement of the nonequilibrium two-particle correlation function by its velocity average; (b) the connection between the nonequilibrium two-point function and the equilibrium three-point function; (c) the link provided by the nonlinear fluctuation-dissipation theorem between the quadratic polarizability and the three-point function.

The equation of state and transport coefficients of strongly coupled plasmas has been an outstanding problem of plasma and many-body physics. The problem, in brief, is to find a method for determining physical characteristics of the system in such a manner that usual perturbation expansion in the "plasma parameter" γ ($\gamma = \kappa^3/4\pi n$, κ^{-1} is the Debye length) can be avoided when γ is not small. Such situations occur in metals and in high-density classical plasmas, e.g., stellar interiors and laser-compressed plasmas. A pioneering idea was put forward by Hubbard,¹ who suggested introducing an effective interaction shielded by the equilibrium pair correlation function. This original approach, however, met with difficulty since it violated structural requirements imposed on the dielectric function by high-frequency conductivity and low-frequency compressibility sum rules. Further progress in the field was obtained by Singwi *et al.*² who put the approximation on a more systematic basis. The main feature of their approach is that the dielectric function is calculated through the first Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) equation in terms of the equilibrium pair correlation function with self-consistency guaranteed by application of the linear fluctuation-dissipation theorem (FDT). In this scheme, however, the equally important, proper nonequilibrium part of the two-particle correlation function is ignored. While Singwi's theory led to substantial improvements in the calculation of the pair correlation function and satisfied the structural require-

ments of the sum rules, it nevertheless failed to reproduce known exact lower-order (in γ) perturbation results. A different method, not based on a self-consistency requirement, but rather on approximating the ternary correlation function by a Debye-like structure, has been followed by Ichimaru.³ His resulting dielectric function does satisfy both the structural requirements of the sum rules and does reproduce the correct perturbation expansion.⁴

The present Letter displays a new self-consistent approach to the problem of strongly coupled plasmas. This approach is made possible by injecting into the approximation scheme a new element, the *nonlinear* FDT derived earlier by the authors.⁵ The essence of the nonlinear FDT is that it connects quadratic response functions to three-point equilibrium correlations. By further relating the three-point equilibrium correlation to two-point nonequilibrium correlations it is possible to formulate the problem in terms of a self-consistent calculation for the combined set of *linear and quadratic* response functions. The merits of this approach are the following: (i) All the requirements listed above are satisfied; (ii) in contrast to other approaches, no artificial assumption is used concerning the structure of the equilibrium or nonequilibrium correlation functions; (iii) since the main approximation appears in approximating the *quadratic* polarizability rather than the linear one, we expect the inaccuracy of the scheme to be thereby reduced; (iv) the scheme provides a full nontrivial extension, without any

further assumption, to the calculation of the dynamical (ω -dependent) polarizability.

The result of this work is displayed in the form of a nonlinear integral equation for the linear dielectric function as the unknown; ultimately this equation has to be solved numerically. The static solution should provide information on the equation of state and possible criteria for gas-liquid phase transitions. From the dynamical solution one should be able to infer the structure, dispersion, and damping of collective modes, and their interaction with particles.

In the following, we outline the simpler static

and more complex dynamical theory. The model we consider is a classical electron plasma embedded in a neutralizing background. The restrictions listed are not essential and extension of the scheme to more general systems is possible and will be discussed elsewhere.

The perturbation of the system by a small external field \vec{E} produces perturbation expansions in the one- and two-particle distribution functions $F(1)$ and $G(12)$. The wave-vector- and frequency-dependent linear dielectric function, $\epsilon(\vec{k}\omega) = 1 + \alpha(\vec{k}\omega)$, is calculated from the perturbed first kinetic equation of the BBGKY hierarchy,

$$-i(\omega - \vec{k} \cdot \vec{v})F'(\vec{k}\omega, \vec{v}_1) - (e/m)\vec{E}'(\vec{k}\omega) \cdot \frac{\partial F^0(\vec{v}_1)}{\partial \vec{v}_1} = \frac{i}{mV} \frac{\partial}{\partial \vec{v}_1} \cdot \sum_{\vec{q}} \vec{q} \varphi(q) \int \mathcal{G}'(\vec{q}, \vec{k} - \vec{q}; \vec{v}_1, \vec{v}_2; \omega) d^3v_2, \quad (1)$$

where $\varphi(q) = 4\pi e^2/q^2$, V is the volume of the system, $\vec{E}'(\vec{k}\omega)$ is the *total* first-order electric field, and $\mathcal{G}(12)$ is the correlation component of $G(12)$. (Prime superscripts here and in the following will label order with respect to the external perturbation.)

The Fourier transform of the two-point correlation $\langle \rho(1)\rho(2) \rangle'$ is related to $\mathcal{G}'(12)$ by

$$\int \int \mathcal{G}'(\vec{q}, \vec{k} - \vec{q}; \vec{v}_1, \vec{v}_2; \omega) d^3v_1 d^3v_2 = e^{-2} \langle \rho_{\vec{k}-\vec{q}} \rho_{\vec{q}} \rangle'(\omega) - n'(\vec{k}\omega) [1 + n_0 V (\delta_{\vec{q}, \vec{\sigma}} + \delta_{\vec{k}, \vec{q}})], \quad (2)$$

where n_0 and n' are the unperturbed and perturbed number densities, and where the $\langle \dots \rangle'$ denotes averaging with respect to the first-order perturbed ensemble.

We further note that by explicitly evaluating $\langle \rho(1)\rho(2) \rangle'$ with the aid of the perturbed Liouville equation, it can be expressed in terms of the equilibrium three-point correlation function $\langle \rho(1)\rho(2)\rho(3) \rangle^0$ through

$$\langle \rho_{\vec{k}-\vec{q}} \rho_{\vec{q}} \rangle'(\omega) = \frac{\beta \hat{E}(\vec{k}\omega)}{Vk} \left\{ \omega \int_0^\infty dt e^{i\omega t} \langle \rho_{\vec{k}-\vec{q}}(0) \rho_{\vec{q}}(0) \rho_{-\vec{k}}(-t) \rangle^0 - \langle \rho_{\vec{k}-\vec{q}}(0) \rho_{\vec{q}}(0) \rho_{-\vec{k}}(0) \rangle^0 \right\}, \quad (3)$$

where β^{-1} is the temperature in energy units. Our principal approximation consists of replacing the two-particle, velocity-dependent correlation function by its *velocity average* [to be referred to as the "velocity average approximation" (VAA)]:

$$\int d^3v_2 \mathcal{G}(\vec{x}_1, \vec{x}_2; \vec{v}_1, \vec{v}_2; t) \approx \frac{F(\vec{x}_1, t, \vec{v}_1)}{n(\vec{x}_1, t)} \int \int d^3v_1' d^3v_2 \mathcal{G}(\vec{x}_1, \vec{x}_2; \vec{v}_1', \vec{v}_2; t). \quad (4)$$

Equations (1), (2), and the perturbed version of (4) now combine to give

$$-i(\omega - \vec{k} \cdot \vec{v})F'(\vec{k}\omega, \vec{v}) - (e/m)\vec{E}'(\vec{k}\omega) \cdot \frac{\partial F^0(\vec{v})}{\partial \vec{v}} = \frac{i}{n_0 V m e^2} \frac{\partial F^0(\vec{v})}{\partial \vec{v}} \cdot \sum_{\vec{q}} \vec{q} \varphi(q) \langle \rho_{\vec{k}-\vec{q}} \rho_{\vec{q}} \rangle'(\omega) - \frac{i n'(\vec{k}\omega)}{m} \frac{\partial F^0(\vec{v})}{\partial \vec{v}} \cdot \vec{k} \varphi(k). \quad (5)$$

Consider first the static ($\omega = 0$) version of the theory. We introduce the static proper three-point function T defined by

$$T(\vec{q}, \vec{k} - \vec{q}) = (2V)^{-1} \langle \rho_{\vec{q}}(0) \rho_{\vec{k}-\vec{q}}(0) \rho_{-\vec{k}}(0) \rangle^0 |_{\vec{q}, \vec{q}-\vec{k}, \vec{k} \neq 0}. \quad (6)$$

Then upon splitting the $\omega = 0$ part of (3) into its improper and proper components and by application of the linear FDT to the former, the static version of (5) becomes

$$F'(\vec{k}, \vec{v}) = - (ie/mk) E'(\vec{k}) \frac{\vec{k} \cdot [\partial F^0(\vec{v})/\partial \vec{v}]}{\vec{k} \cdot \vec{v}} - \frac{2i\beta \hat{E}(k)}{V n_0 e^2 m k} \frac{\varphi(k) \vec{k} \cdot [\partial F^0(\vec{v})/\partial \vec{v}]}{\vec{k} \cdot \vec{v}} \sum_{\vec{q}} \frac{\vec{k} \cdot \vec{q}}{q^2} T(\vec{q}, \vec{k} - \vec{q}). \quad (7)$$

Equation (7) expresses the important fact that in the VAA the exact *nonequilibrium two-particle* corre-

lation function is replaced by the *equilibrium three-particle* correlation function. Then from (7) and the nonlinear FDT,⁵

$$T(\vec{q}, \vec{k} - \vec{q}) = \frac{ikq|\vec{k} - \vec{q}|}{4\pi\beta^2} \frac{\alpha(\vec{q}, \vec{k} - \vec{q})}{\epsilon(\vec{q})\epsilon(\vec{k} - \vec{q})\epsilon(\vec{k})}, \quad (8)$$

for the static quadratic polarizability, $\alpha(\vec{q}, \vec{k} - \vec{q})$, one ultimately obtains the desired equation for the static linear polarizability:

$$\alpha(\vec{k}) = \alpha_0(k)[1 + r(\vec{k})], \quad (9)$$

where $\alpha_0(k) = 4\pi n_0 \beta e^2 / k^2 \equiv \kappa^2 / k^2$ is its Vlasov value, and

$$r(\vec{k}) = \frac{2i}{\beta n_0 e V} \sum_{\vec{q}} \frac{(\vec{k} \cdot \vec{q})|\vec{k} - \vec{q}|}{kq} \frac{\alpha(\vec{q}, \vec{k} - \vec{q})}{\epsilon(\vec{q})\epsilon(\vec{k} - \vec{q})}. \quad (10)$$

(Subscripts here and in the following refer to ordering in the coupling parameter γ .)

Further progress now is contingent upon the approximation to be used to evaluate $\alpha(\vec{q}, \vec{k} - \vec{q})$. Various options are possible; these, however, will not be discussed here. We consider only the simplest procedure, which is to adopt the Vlasov approximation for $\alpha(\vec{q}, \vec{k} - \vec{q})$, viz.,^{5,6}

$$\alpha(\vec{q}, \vec{k} - \vec{q}) = \alpha_0(\vec{q}, \vec{k} - \vec{q}) \equiv \frac{2\pi i \beta^2 n_0 e^3}{kq|\vec{k} - \vec{q}|}. \quad (11)$$

Equations (9)–(11) constitute the final result of the static theory. The resulting integral equation featuring $\epsilon(\vec{k})$ as the unknown has a fairly simple structure. An analysis of the solutions of this equation will be considered at a later time.⁷

The expression (9) for $\alpha(\vec{k})$ structurally satisfies the compressibility sum rule, $\alpha(\vec{k} \rightarrow 0) = A(\kappa^2/k^2)$, for the $\vec{k} \rightarrow 0$ limit of $r(\vec{k})$ is found to be a bounded sum. Moreover, to lowest order in γ , $r(\vec{k} \rightarrow 0)$ tends to the required value³ $(A - 1) = \gamma/4$.

Application of the linear FDT to (9) yields the corresponding equilibrium pair correlation g . In the weak-coupling limit, its expansion in powers of γ , i.e., $g = g_1 + g_2 + \dots$, verifies that g_2 is indeed the correct O'Neil-Rostoker⁸ expression.

We now present results of the more general dynamical version of the approximation scheme. Introducing the dynamical proper triplet correlation function T , defined by

$$T(\vec{q}\omega'; \vec{k} - \vec{q}, \omega'') \delta(\omega' + \omega'' - \omega) = (4\pi V)^{-1} \langle \rho_{\vec{q}}(\omega') \rho_{\vec{k}-\vec{q}}(\omega'') \rho_{-\vec{k}}(-\omega) \rangle^0_{\vec{q}, \vec{q}-\vec{k}, \vec{k} \neq 0}, \quad (12)$$

we obtain the corresponding ($\omega \neq 0$) generalization of (9) in the form

$$\frac{\alpha(\vec{k}\omega)}{\epsilon(\vec{k}\omega)} = \frac{\alpha_0(\vec{k}\omega)}{\epsilon_0(\vec{k}\omega)} [1 + w(\vec{k}\omega)], \quad (13)$$

$$w(\vec{k}\omega) = \frac{4\pi\beta}{n_0 e k^2 V} \sum_{\vec{q}} \frac{\vec{k} \cdot \vec{q}}{q^2} \left[\frac{i\omega}{\pi} \int_{-\infty}^{\infty} d\omega' \int_{-\infty}^{\infty} d\omega'' \delta_+(\omega - \omega' - \omega'') T(\vec{q}\omega'; \vec{k} - \vec{q}, \omega'') + 2T(\vec{q}, \vec{k} - \vec{q}) \right]. \quad (14)$$

[Note that $r(\vec{k}) = \epsilon(\vec{k})w(\vec{k}\omega = 0)$.] From the dynamical nonlinear FDT,⁵

$$T(\vec{q}\omega'; \vec{k} - \vec{q}, \omega'') = \frac{kq|\vec{k} - \vec{q}|}{2\pi\beta^2 \epsilon(\vec{q}\omega') \epsilon(\vec{k} - \vec{q}, \omega'') \epsilon(\vec{k}, \omega' + \omega'')} \times \text{Im} \left[\frac{\alpha(\vec{q}\omega'; \vec{k} - \vec{q}, \omega'')}{\omega' \omega''} - \frac{\alpha(-\vec{q}, -\omega'; \vec{k}, \omega' + \omega'')}{\omega'(\omega' + \omega'')} - \frac{\alpha(\vec{k}, \omega' + \omega''; \vec{q} - \vec{k}, -\omega'')}{\omega''(\omega' + \omega'')} \right], \quad (15)$$

$w(\vec{k}\omega)$ can then be expressed entirely in terms of linear and quadratic polarizabilities. Here again we adopt the simplest Vlasov approximation⁶ for the latter. Equations (13)–(15) constitute the final result of the dynamical theory. The resulting nonlinear integral equation includes frequency integrals and is much more involved than its static counterpart. This is a consequence of the dynamical $r(\vec{k}\omega) = \epsilon(\vec{k}\omega) \times w(\vec{k}\omega)$ being structurally different from the static $r(\vec{k})$, and is a feature not properly accounted for in other theories.^{2,3} Analysis of solutions of (13) to (15) will again be deferred to later publications.

Concerning the high-frequency behavior of Eqs. (13) and (14), we find that

$$\epsilon(\vec{k}\omega \rightarrow \infty) = 1 - \frac{\omega_p^2}{\omega^2} - \frac{\omega_p^2}{\omega^4} [k^2 a^2 - s(\vec{k})], \quad (16)$$

$$s(\vec{k}) = -\frac{1}{\beta n_0 m k^2 V} \sum_{\vec{q}} \frac{(\vec{k} \cdot \vec{q})^2}{q^2} \left[|\vec{k} - \vec{q}|^2 \frac{\alpha(\vec{k} - \vec{q})}{\epsilon(\vec{k} - \vec{q})} - q^2 \frac{\alpha(\vec{q})}{\epsilon(\vec{q})} \right], \quad (17)$$

where $a^2 = 3/(\beta m)$. Equation (16) verifies the conductivity sum rule. Note that to lowest order in γ , $s(\vec{k} \rightarrow 0) = \frac{11}{45} \gamma k^2 a^2$. This suggests the possibility that, for γ sufficiently large, the slope of the plasma oscillation dispersion curve changes sign, accompanied by the appearance of a low-frequency electron sound mode.

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⁷A computer analysis of Eqs. (9)–(11) is underway. The convergence of an iteration scheme is indicated for at least $\gamma \leq 5$.

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Faraday Rotation of Small Polarons*

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The Faraday rotation of small polarons is investigated. For three-site hopping the rotation angle has maxima located symmetrically about the frequency at which there is a maximum in the optical absorption. For four-site hopping, the rotation angle is found to exhibit an anomalous change in sign. Experimentally determined parameters for rutile (TiO₂) suggest that the anomalous four-site effect is observable.

Friedman and one of the present authors have calculated the dc Hall mobility of small polarons by using two techniques: an intuitive jump-probability approach^{1,2} and a Kubo linear-response approach.³ Here we have calculated the real part of the antisymmetric part of the conductivity tensor at *nonzero* frequency using the Kubo formulation. In the region of low absorption this gives the angle of rotation of the plane of polarization of light which traverses small-polaron material in the direction of a constant magnetic field,⁴ i.e., the Faraday rotation of small polarons.

It has long been recognized that the optical absorption due to small polarons can be understood

in terms of the Franck-Condon principle.^{5,6} At the occupied site, the electron polarizes the lattice in such a way that it requires an energy of $2E_b$ (where E_b is the polaron binding energy) to make a "vertical" transition to a similar state at another lattice site. The other sites may then be regarded as being at an energy $\hbar\omega_m = 2E_b$ above the occupied site. This defines the energy levels. The existence of these levels is exploited in the calculation of optical properties by noting that at frequencies comparable with the absorption frequency ω_m , by far the largest contribution to the antisymmetric conductivity will arise from virtual transitions in which the small polaron ab-