Real-Time Kadanoff-Baym Approach to Plasma Oscillations in a Correlated Electron Gas

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A nonequilibrium Green's functions approach to the collective response of correlated Coulomb systems at finite temperatures is presented. It is shown that solving Kadanoff-Baym-type equations of motion for the two-time correlation functions including the external perturbing field allows one to compute the plasmon spectrum with collision effects in a systematic and consistent way. The scheme has a "built-in" sum-rule preservation and is simpler to implement numerically than the equivalent equilibrium approach based on the Bethe-Salpeter equation.

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The dynamic properties and the plasmon spectrum of Coulomb systems continue to attract the interest of researchers in many fields, in particular, condensed matter theory, e.g., [1–4], plasma physics, e.g., [5–7], and electronic bilayer liquids [8]. This is due to the fact that the density response to an external perturbation, given, e.g., by the dynamic structure factor $S(q, \omega)$, is a sensitive indicator of the state of a charged particle system which can be directly measured in x-ray or light scattering and electron-energy-loss experiments. This is particularly valuable for strongly coupled many-body systems, such as dense plasmas, metals, or semiconductors at low temperature.

In recent years there has been considerable progress in the theoretical account of the impact of correlations among the carriers on collective excitations, i.e., in theories which go beyond the mean-field level [time-dependent Hartree/Vlasov or random phase approximation (RPA)]. It is now commonly accepted that any model has to obey certain consistency requirements which, in particular, are related to the preservation of sum rules for the inverse dielectric function; see Refs. [1–4] for a discussion. Among the successful approaches, we mention attempts to construct local field corrections (see references in [4]), kinetic theory concepts to incorporate collisions into the dielectric function, e.g., [9], and Green's functions methods originally developed by Baym and Kadanoff [10] and others. The latter approach is of particular interest as it allows for a systematic first principles treatment of correlations, and sum-rule preservation is easily guaranteed by using so-called *conserving* approximations for the Green's functions [4,10,11].

In most Green's functions treatments [1–4,10,11], linear response theory is used which relates the density response function to the retarded *one-particle–one-hole Green's function of the unperturbed system*, the calculation of which is the central problem. This task is accomplished by solving a Bethe-Salpeter equation (BSE), the quality of the results being determined by the choice of the four-point particle-hole-irreducible (PHI) vertex K. While high-level approximations for K have been investigated for metals at zero temperature [4,11], *finite-temperature* treatments are restricted to much simpler approximations [2]. Moreover, the collective response from a *nonequilibrium state*, which is of high interest, e.g., in laser excited semiconductors or laser plasmas, is completely out of reach in the BSE approach.

In this Letter, we present a scheme which allows (i) the computation of the linear response at finite temperature fully including vertex corrections, (ii) the nonlinear response to a strong perturbation, and (iii) the response from an arbitrary nonequilibrium state. Our approach is based on directly computing the time-dependent density fluctuations of the electron gas under an external perturbing field, from which we obtain the density response function. We calculate the *nonequilibrium* (two-time) *one-particle* Green's functions by solving a generalized Kadanoff-Baym-type equation with the external perturbing field included. If the unperturbed system is in equilibrium, sum rules are again preserved by adopting conserving approximations of the self-energy [10].

There is a one-to-one equivalence between levels of approximations in the two approaches. For each choice of the approximate self-energy in the present approach, its formal functional derivative with respect to the one-particle Green's function gives the equivalent PHI vertex in the Bethe-Salpeter approach. However, in investigations involving more sophisticated approximations, our approach has the advantage that the self-energies are formally much simpler, and hence easier to implement numerically, than their equivalent PHI vertices. We underline that this efficient and conserving calculational scheme is not limited to the problem of plasma oscillations but is of interest also for current studies of finite temperature spin modes in Fermi liquids [12] and finite temperature collective excitations in nuclei [13]. In the following, to avoid confusion in terminology, we append the suffix "BS" to the labels of the approximations in the Bethe-Salpeter approach.

We consider a correlated electron gas in a neutralizing background under the influence of an external potential U described by the Hamiltonian $\hat{H} = \hat{H}_{\rm sys} + \hat{H}_{\rm ext}$, with

$$\hat{H}_{\rm sys} = \sum_{\mathbf{k}} \epsilon_{\mathbf{k}} a^{\dagger}_{\mathbf{k}} a_{\mathbf{k}} + \frac{1}{2} \sum_{\mathbf{k}_1 \mathbf{k}_2, \mathbf{q} \neq 0} V(q) a^{\dagger}_{\mathbf{k}_1 + \mathbf{q}} a^{\dagger}_{\mathbf{k}_2 - \mathbf{q}} a_{\mathbf{k}_2} a_{\mathbf{k}_1} ,$$
$$\hat{H}_{\rm ext} = \sum_{\mathbf{q}} U(-\mathbf{q}, t) \sum_{\mathbf{k}} a^{\dagger}_{\mathbf{k}+\mathbf{q}} a_{\mathbf{k}} . \quad (1)$$

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Here k, q are momenta, ϵ_k is the one-particle energy, and $V(q)$ and $U(q)$ are the spatial Fourier components of the Coulomb potential and the external potential, respectively. $a^\dagger$ (a) are Heisenberg creation (annihilation) operators evolving with the total Hamiltonian $\hat{H}$. (Spin degrees of freedom are not of interest for our analysis, so spin indices will be suppressed.) The nonequilibrium properties of the inhomogeneous electron gas are defined by the two-time correlation functions

$$\begin{aligned} G^<(\mathbf{k}+\mathbf{q},t_1;\mathbf{k},t_2) &= i\langle a^\dagger_{\mathbf{k}}(t_2)a_{\mathbf{k}+\mathbf{q}}(t_1)\rangle; \\ G^>(\mathbf{k}+\mathbf{q},t_1;\mathbf{k},t_2) &= -i\langle a_{\mathbf{k}+\mathbf{q}}(t_1)a^\dagger_{\mathbf{k}}(t_2)\rangle, \end{aligned} \tag{2}$$

where the statistical averaging is over the density operator of the initial state. In particular, the density is given by $n(\mathbf{q},t) = -i\sum_{\mathbf{k}} G^<(\mathbf{k}+\mathbf{q},t;\mathbf{k},t)$. The time evolution of $G^>$ and $G^<$ is governed by the Kadanoff-Baym equations (KBE) [10],

$$\left(i\hbar\frac{\partial}{\partial t_1} - \epsilon_{\mathbf{k}_1}\right)G^{\gtrless}(\mathbf{k}_1t_1;\mathbf{k}_2t_2) = \sum_{\mathbf{q}} U(-\mathbf{q},t_1)G^{\gtrless}(\mathbf{k}_1-\mathbf{q},t_1;\mathbf{k}_2t_2) + \sum_{\bar{\mathbf{k}}} \Sigma^{\mathrm{HF}}(\mathbf{k}_1t_1;\bar{\mathbf{k}}t_1)G^{\gtrless}(\bar{\mathbf{k}}t_1;\mathbf{k}_2t_2) + I^{\gtrless}(\mathbf{k}_1t_1;\mathbf{k}_2t_2) \tag{3}$$

(to be supplemented with the adjoint equation), where Σ^{HF} is the Hartree-Fock self-energy, and the collision integrals $I^{\gtrless}$ contain the short-range correlation effects (see below).

As we are interested in the dynamical response of the electron gas to a longitudinal electrostatic perturbation, we choose $U(\mathbf{q},t) = U_0(t)\delta_{\mathbf{q},\mathbf{q}_0}$. Before the onset of the field, $t < t_0$, the system is homogeneous, $G^{\gtrless}(\mathbf{k}_1t_1;\mathbf{k}_2t_2) \sim \delta_{\mathbf{k}_1,\mathbf{k}_2}$; however, for $t > t_0$, the field gives rise to harmonic modulations

$$G^{\gtrless}_{\mu_1,\mu_2}(\mathbf{k}t_1t_2) \equiv G^{\gtrless}(\mathbf{k}+\mu_1\mathbf{q}_0,t_1;\mathbf{k}+\mu_2\mathbf{q}_0,t_2), \tag{4}$$

where μ_1 and μ_2 are integers running from $-\infty$ to ∞. q_0 enters G_{μ_1,μ_2} as a parameter and will be omitted. Because of the symmetry properties $G^{\gtrless}_{\mu_1+n,\mu_2+n}(\mathbf{k}t_1t_2) = G^{\gtrless}_{\mu_1\mu_2}(\mathbf{k}+n\mathbf{q}_0,t_1t_2)$ and $G^{\gtrless}_{\mu_1,\mu_2}(\mathbf{k}t_1t_2) = -G^{\gtrless *}_{\mu_2,\mu_1}(\mathbf{k}t_2t_1)$, only the matrix elements $G^{\gtrless}_{n0}, n = 0,\pm1,\pm2,\ldots$ are independent. As a result, Eq. (3) transforms into a system of equations for the functions $G^{\gtrless}_{n0}$, i.e., one of the two momentum arguments of $G^{\gtrless}$ in Eq. (3) has been replaced by the discrete "level" index n. Obviously, this representation closely resembles the multilevel/multiband kinetic equations (Bloch equations) familiar from atomic or semiconductor optics, if written in terms of two-time correlation functions, e.g., [14]. Only here, G_{00} corresponds to the spatially homogeneous state, while $G^<_{n0}$ describes transitions of an electron from the momentum state $\mathbf{k}+n\mathbf{q}_0$ at $t = t_1$ into state $\mathbf{k}$ at $t = t_2$; cf. definition (2). In particular, the equal-time components of $G^<_{00}$ and $G^<_{m0}$ yield, respectively, the homogenous density component $n_0(t) = -i\sum_{\mathbf{k}} G^<_{00}(\mathbf{k}tt)$ and the field-induced fluctuations

$$\delta n(\mathbf{q},t) = \sum_{m\neq0}\delta n_m = -i\sum_{m\neq0}\delta_{\mathbf{q},m\mathbf{q}_0}\sum_{\mathbf{k}} G^<_{m0}(\mathbf{k}tt). \tag{5}$$

In situations where a perturbation treatment of the external field is applicable, the leading order of the Fourier components of the density is $\delta n_m \sim O(U_0^m)$. Since the main subject of our paper is the effect of correlations (collisions) on the plasmon spectrum, we will focus on the weak-field (linear response) limit below. Then, we neglect all components of $G^{\gtrless}$ except $G^{\gtrless}_{00}$ and $G^{\gtrless}_{10}$. Up to first order in the field, the equations for $G^{\gtrless}_{10}$ read, for any fixed $\mathbf{q}_0$ (summation over $m = 0, 1$ and integration over $\bar{t}$ from $-\infty$ to ∞ is implied),

$$\begin{aligned} \left(i\hbar\frac{\partial}{\partial t_1} - \epsilon_{\mathbf{k}+\mathbf{q}_0}\right)G^{\gtrless}_{10}(\mathbf{k}t_1t_2) &= U_0(t_1)G^{\gtrless}_{00}(\mathbf{k}t_1t_2) + \Sigma^{\mathrm{HF}}_{1m}(\mathbf{k}t_1)G^{\gtrless}_{m0}(\mathbf{k}t_1t_2) \\ &\quad + \Sigma^R_{1m}(\mathbf{k}t_1\bar{t})G^{\gtrless}_{m0}(\mathbf{k}\bar{t}t_2) + \Sigma^{\gtrless}_{1m}(\mathbf{k}t_1\bar{t})G^A_{m0}(\mathbf{k}\bar{t}t_2), \\ \left(-i\hbar\frac{\partial}{\partial t_2} - \epsilon_{\mathbf{k}}\right)G^{\gtrless}_{10}(\mathbf{k}t_1t_2) &= U_0(t_2)G^{\gtrless}_{11}(\mathbf{k}t_1t_2) + G^{\gtrless}_{1m}(\mathbf{k}t_1t_2)\Sigma^{\mathrm{HF}}_{m0}(\mathbf{k}t_2) \\ &\quad + G^R_{1m}(\mathbf{k}t_1\bar{t})\Sigma^{\gtrless}_{m0}(\mathbf{k}\bar{t}t_2) + G^{\gtrless}_{1m}(\mathbf{k}t_1\bar{t})\Sigma^A_{m0}(\mathbf{k}\bar{t}t_2), \end{aligned} \tag{6}$$

whereas $G^{\gtrless}_{00}$ obey the "conventional" spatially homogeneous field-free equations. In Eq. (6), $G^{\gtrless}_{11}(\mathbf{k}t_1t_2) \equiv G^{\gtrless}_{00}(\mathbf{k}+\mathbf{q}_0,t_1t_2)$, the retarded and advanced functions are defined by (F denotes G or Σ)

$$F^{R/A}_{\lambda_1\lambda_2}(\mathbf{k}t_1t_2) = \pm\Theta[\pm(t_1-t_2)]\times\{F^>_{\lambda_1\lambda_2}(\mathbf{k}t_1t_2) - F^<_{\lambda_1\lambda_2}(\mathbf{k}t_1t_2)\}, \qquad \lambda_{1,2} = 0,1, \tag{7}$$

and the Hartree-Fock self-energy is

$$\Sigma^{\mathrm{HF}}_{\lambda0}(\mathbf{k}t) = \delta_{\lambda1}V(q_0)\sum_{\mathbf{p}}(-i)G^<_{10}(\mathbf{p}tt) - \sum_{\mathbf{p}}(-i)G^<_{\lambda0}(\mathbf{k}-\mathbf{p},tt)V(p), \qquad \lambda = 0,1. \tag{8}$$

The self-energies $\Sigma_{10}^{\gtrless}$ and $\Sigma_{10}^{R/A}$ are of first order in the field and are obtained from the respective "00" components by replacing one G_{00} at a time by G_{10} and summing over all terms generated this way, and Σ_{11} follows from Σ_{00} by replacing G_{00} by G_{11}. The equations for G_{00} and G_{10} are to be supplemented by the proper initial conditions for $G_{00}^{\gtrless}$ corresponding to a correlated spatially homogeneous electron gas; see below. Furthermore, $G_{10}(\mathbf{k}t_0t_0) \equiv 0$.

Let us now consider how the dielectric linear response functions can be determined from the solution of Eqs. (6) and what their properties are. Using Eq. (5), we find in linear response

$$\sum_{\mathbf{k}} G_{10}^{<}(\mathbf{k}tt) = i\delta n_{\mathbf{q}_0}(t) = \int_{-\infty}^{\infty} d\bar{t}\, \chi^R(\mathbf{q}_0, t, \bar{t}) U_0(\bar{t}), \tag{9}$$

where χ^R is a retarded susceptibility which, in general, depends on two times. If the unperturbed system is in a stationary state, $\chi^R(t,\bar{t}) \to \chi^R(t-\bar{t})$, allowing one to apply the convolution theorem to Eq. (9) with the result (ω dependence denotes the Fourier component)

$$\chi^R(\mathbf{q}_0, \omega) = \frac{\sum_{\mathbf{k}} G_{10}^{<}(k,\omega)}{U_0(\omega)}, \tag{10}$$

which immediately yields the retarded inverse dielectric function and the dynamic structure factor

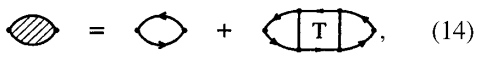
$$\epsilon^{R-1}(\omega, \mathbf{q}_0) = 1 + \frac{V(q_0)}{U_0(\omega)} \sum_{\mathbf{k}} G_{10}^{<}(\mathbf{k},\omega), \tag{11}$$

$$S(\omega, \mathbf{q}_0) = -\frac{1}{\pi n_0 U_0(\omega)} \sum_{\mathbf{k}} \mathrm{Im} G_{10}^{<}(\mathbf{k},\omega). \tag{12}$$

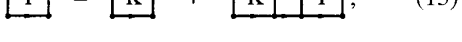
Now, the quality of the plasmon spectrum (12) computed from $G_{10}(t_1t_2)$ is fully determined by the approximation for the *field-free self-energies* Σ_{00} in Eqs. (6). In particular, if $\Sigma_{00}^{\mathrm{HF}} = \Sigma_{00}^{\gtrless} = 0$ (noninteracting electrons gas), Eq. (10) reduces to the familiar Lindhard polarization, $\chi^R \equiv \Pi^{R0}$. Further, if only the Hartree mean field is included, $\Sigma_{00}^{\mathrm{HF}} \equiv \Sigma_{00}^{\mathrm{H}}$, one recovers the RPA-BS result (full ring sum), $\chi^R = \frac{\Pi^{R0}}{1 - V\Pi^{R0}}$, or, equivalently,

$$\chi^R = \chi^* + \chi^* V \chi^R, \tag{13}$$

with $\chi^* \equiv \Pi^{R0}$. Finally, with the Fock and correlation terms, Σ^{F} and $\Sigma_{00}^{\gtrless}$, included also, one again recovers Eq. (13), but with a more general expression for χ^*, the proper (irreducible) polarization:



(14)



(15)

where Eq. (14) starts with Π^{R0} (first diagram) but now contains exchange and correlation corrections (second diagram) in terms of the particle-hole T matrix T which obeys the Lippmann-Schwinger equation (15) with the general interaction kernel K (see below). One readily recognizes in Eq. (13) the familiar *field-free Bethe-Salpeter equation* which thus is a direct consequence of the *Kadanoff-Baym equations with weak external field* (6). We underline that this result applies to equilibrium and *arbitrary nonequilibrium* situations (notice that all derivations are performed on the Keldysh contour, and directed lines denote *full* Green's functions with self-energy insertions) [15].

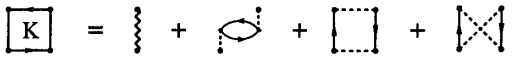
As we demonstrate below, this connection between the BS and KB approaches is particularly fruitful for the dielectric response of a correlated electron gas: (i) as the BS approach is a standard formalism for the investigation of correlation effects, e.g., [7], it can be used to classify approximations and their properties; (ii) there exists a one-to-one correspondence between the self-energies $\Sigma_{00}^{\gtrless}$ and the PHI vertex K in Eq. (14); (iii) based on the internal consistency of the Kadanoff-Baym formalism, the properties of the plasmon spectrum are completely determined by the approximation for $\Sigma_{00}^{\gtrless}$: in particular, density conservation of $\Sigma_{00}^{\gtrless}$ (which is trivial to meet) guarantees satisfaction of the f sum rule [4].

A valuable practical advantage of the present scheme is that simple approximations for $\Sigma_{00}^{\gtrless}$ correspond to rather complex approximations for K which allows for efficient computation of the plasmon spectrum of correlated systems by solving the KBE (6). We demonstrate this below on the example of the (density conserving) second Born approximation

$$\Sigma_{00}^{\gtrless}(\mathbf{k}tt') = i \sum_{\mathbf{p}} |V_{st}(\mathbf{p})|^2 \Pi_{00}^{\gtrless}(\mathbf{k}-\mathbf{p}, tt') G_{00}^{\lessgtr}(\mathbf{p}, t't)$$

(16)

where V_{st} is the statically screened Coulomb potential (dashed lines in the diagram) and $\Pi_{00}^{\gtrless}$ is the nonequilibrium generalization of the Lindhard polarization bubble, $\Pi_{00}^{\gtrless}(\mathbf{k}tt') = -i\sum_{\mathbf{p}} G_{00}^{\gtrless}(\mathbf{k}+\mathbf{p}, tt') G_{00}^{\lessgtr}(\mathbf{p}t't)$. Proceeding as in Ref. [15], the simple correlation self-energy (16), together with the Fock mean field, transforms into the following PHI vertex K in the BSE:



(17)

and contains contributions from particle-hole (unscreened) Coulomb scattering (first diagram, zigzag line denotes bare Coulomb potential V), excitation of a particle-hole pair (second diagram), and scattering between two particle-hole pairs (last two diagrams). Comparison of Eqs. (16) and (17) reveals the familiar relation between Σ and K: $K = V + \delta\Sigma_{00}/\delta G$ [7,10].

While it is very difficult to solve the BSE with kernel (17) without further simplifying approximations, solving Eqs. (6) with the self-energy Eq. (16) and the Σ_{10} derived from it is quite straightforward. We performed numerical solutions for a strongly correlated electron gas in equilibrium using the numerical procedure which was developed before for the two-time semiconductor Bloch equations

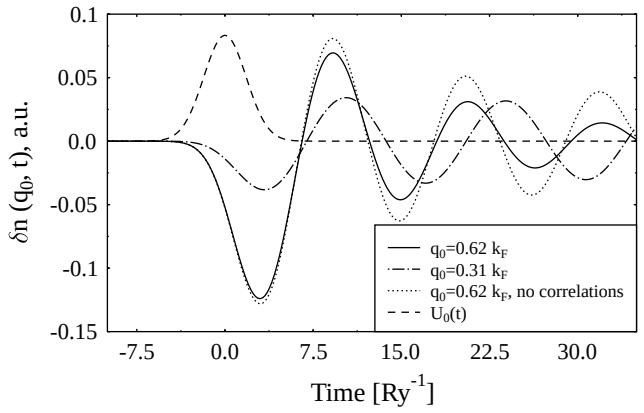


FIG. 1. Density fluctuation of a strongly correlated electron gas for two wave numbers. For comparison, the uncorrelated response for one wave number (dotted line) and the exciting field (dashes) are shown, too. k_F denotes the Fermi momentum, Ry = 13.6 eV.

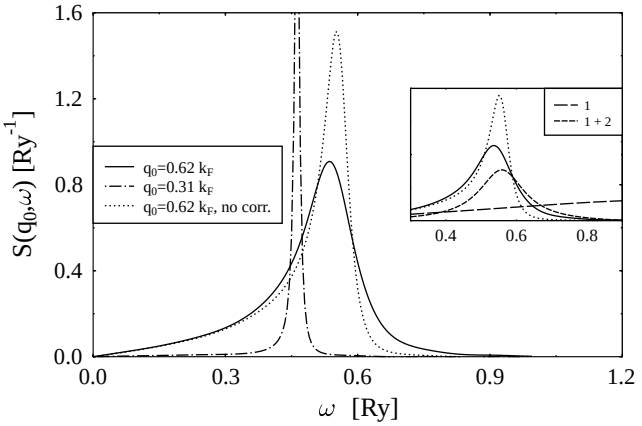


FIG. 2. Dynamic structure factor (12) for the correlated electron gas of Fig. 1 (same line styles). Inset shows S for $q_0 = 0.62k_F$ and contains two other approximations to the correlations corresponding to retaining the first diagram in Eq. (17) and first plus second diagrams, respectively.

[14]. To create a correlated initial state, we run the field-free program for a time longer than the correlation time starting from an uncorrelated distribution. After this, the field U was turned on, where we chose a pulse shape for $U_0(t)$ broad enough to cover the plasmon spectrum. The thus excited density fluctuation is shown in Fig. 1 for a 3D electron gas with Brueckner parameter $r_s = 4$ and temperature $k_BT = 0.69E_F$ (Fermi energy) for two wave numbers. For comparison, also the uncorrelated result is shown (Hartree-Fock self-energies only which is equivalent to RPA-BS plus exchange). While $\delta n(t)$ depends on the explicit form of $U_0(t)$, obviously the linear response quantities $\epsilon^R(\omega)$ and $S(\omega)$, Eqs. (11) and (12), are independent of U_0. Figure 2 shows the dynamic structure factors, corresponding to the results in Fig. 1. Clearly, one sees that the short-range correlations lead to a damping of $\delta n(t)$ in excess of the collisionless Landau damping (cf. Fig. 1), which corresponds to a redshift and an additional broadening of the plasmon peak in the structure factor (Fig. 2). Remarkably, our numerical scheme preserves the f sum rule for the small (large) wave number to 0.03% (0.6%). In contrast, neglecting terms in K but keeping the self-energy insertions in G lead to violation of the sum rule. For example, for inclusion of the first two diagrams only (curve "1 + 2" in the inset in Fig. 2) and the first diagram only ("1") the corresponding numbers are, respectively, 2.1% (0.8%) and 1346% (416%), and for still smaller q_0 the error increases rapidly [3].

In summary, we have presented a new self-consistent approach to the dielectric properties of a correlated electron gas. Using an "interband" generalization and solving the problem in the time domain allows one to take maximum advantage of the self-consistent Kadanoff-Baym scheme: simple approximations for the collision integrals transform into complex correlation corrections in the plasmon spectrum with "built-in" sum-rule preservation. This scheme is straightforwardly extendable to higher order correlations. Moreover, it applies to arbitrary *nonequilibrium* situations and is easily generalized to the *nonlinear* dielectric response in strong fields.

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