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Dielectric function and thermodynamic properties of jellium in the GW approximation.

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The fully self-consistent GW approximation is an established method for electronic structure calculations. Its most serious deficiency is known to be an incorrect prediction of the dielectric response. In this work we examine the GW approximation for the homogeneous electron gas and find that problems with the dielectric response are drastically improved by enforcing the particle-number conservation law in the polarization function. We also find that previously reported data for the ground-state energy were contradicting each other well outside of reported error bounds. Some of these results created a false impression of how accurate the fully self-consistent GW approximation is. Our two independent implementations of the *GW* method agree with the data plotted in Ref. [15], thus confirming only that data set. We also present values for other key Fermi-liquid properties.

Accurately solving the many-electron Schrödinger equation for real solid-state systems is a major challenge of great technological importance. Among available theoretical approaches, approximations based on diagrammatic many-body perturbation theory [1, 2] are attractive because even at low order these approximations can grasp the essential physics and allow to deal with the long-range Coulomb interaction in the thermodynamic limit. The widely used random-phase approximation (RPA), for example, can qualitatively explain features of real metals such as screening, plasmon and Friedel oscillations. In principle, the skeleton diagrammatic expansion allows one to systematically improve on these results and obtain accurate solutions to the many-electron problem. In practice, however, progress is hindered because more sophisticated lowest-order diagrammatic approximations can lead to worse results, while a systematic evaluation of the skeleton series seems computationally too expensive within the conventional implementation (apart from questions about series convergence).

The most widely used diagrammatic method for electronic structure calculations is the so-called GW approximation [3–5]. While clearly going beyond regular RPA by evaluating "bubble"-diagrams in a self-consistent way, the GW approximation has an additional advantage of being a conserving approximation (with respect to the relation between the particle density n and Fermi momentum k_F) as shown by Kadanoff and Baym [6, 7]. It has been established, however, that the GW approximation fails to reproduce some key results for the two-particle correlation functions and does not even properly describe the plasmon properties, in contrast to RPA. This drawback has been clearly demonstrated for a homogeneous electron gas (jellium model) by Holm and von Barth in Ref. [8]. Moreover, incorrect screening properties are expected to have a feedback on single-particle spectra of real materials for which the GW approximation sometimes fails to account for the observed value of the absolute band-gap [9].

In this article we present a simple strategy to restore the physical two-particle correlation properties within the conventional GW approximation. Our trick can be applied at every order of the skeleton expansion, and does not produce any systematic bias in the infinite-order limit for convergent series. It could therefore be used in the future within a Diagrammatic Monte Carlo approach [10–12]. We focus here on the jellium model, describing Coulomb-interacting electrons moving against a positively charged uniform background.

We also found that previously published results for the ground-state energy per particle E/N obtained with the standard GW approach were in strong (well outside of reported error bounds) disagreement with each other, see Refs. [8, 13–15]. We have carefully developed two independent GW codes, which both confirm the ground-state energy data plotted in Ref. [15]. We provide accurate values for the ground-state energy, the quasiparticle Z-factor, and the effective mass renormalization m_*/m (where m is the bare electron mass) at the Fermi level.

Formalism. Let us start by briefly reviewing the GW approximation. It is based on the lowest-order skeleton diagrams for the irreducible self-energy Σ_σ (σ is the spin index) and the irreducible polarization Π. In the position-imaginary time (r,τ)-representation it reads:

$$\Sigma_\sigma(r,\tau) = -G_\sigma(r,\tau)W(r,-\tau)\,, \tag{1}$$

$$\Pi(r,\tau) = \sum_\sigma G_\sigma(r,\tau)G_\sigma(r,-\tau)\,, \tag{2}$$

where G_σ is the one-body Green's function and W the effective screened interaction. These are self-consistently

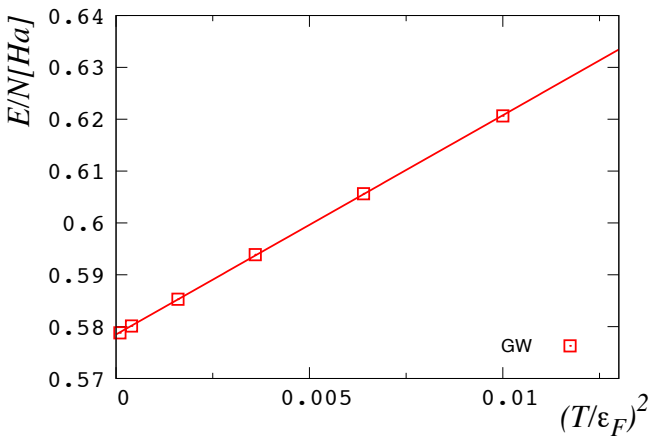

FIG. 1. Energy per electron (in Hartrees) as a function of $(T/\epsilon_F)^2$ revealing the Fermi-liquid behavior. The solid line is a linear fit giving a ground-state energy of $E/N = 0.5783(2)$ Ha.

defined through solutions of the Dyson equations in the momentum-Matsubara frequency (k, ω_n)-representation:

$$G_\sigma(k,\omega_n)^{-1} = G^0_\sigma(k,\omega_n)^{-1} - \Sigma_\sigma(k,\omega_n)\ , \qquad (3)$$

$$W(k,\omega_n)^{-1} = V(k)^{-1} - \Pi(k,\omega_n)\ , \qquad (4)$$

where $V(k) = 4\pi e^2/k^2$ is the bare Coulomb interaction and G^0_σ is the free one-body Green's function. Knowing the one-body Green's function G is sufficient for obtaining the system's energy, as well as quasiparticle properties such as m_* and Z, see Refs. [1, 2]. We performed all calculations at finite temperatures well below the Fermi energy ϵ_F. For ground-state properties we extrapolated results to zero temperature using the Fermi-liquid behavior. In Fig. 1 we show a typical plot for energy at $r_s = 1$, with standard definition of r_s as the ratio of the typical inter-particle spacing and the Bohr radius.

Dielectric response. The work by Holm and von Barth [8] has established that the GW approximation leads to unphysical behaviour of the two-particle correlation functions. More precisely, it was found that the spectral function $S(k,\omega)$ of the irreducible polarization has incorrect behavior at frequencies $\omega > kv_F$, where v_F is the Fermi velocity; as a consequence, the real part of the dielectric function $\epsilon(k,\omega) = 1 - (4\pi e^2/k^2)\Pi(k,\omega)$ at small momenta $k \ll k_F$ has its zero shifted away from the plasmon frequency $\omega_p = \sqrt{4\pi n e^2/m}$ to completely unphysical values, see Fig. 3 in Ref. [8].

Our results agree with this key observation: we also find that at $k \ll k_F$ and $\omega_n \gg kv_F$ the irreducible polarization is *orders of magnitude* larger than the expected values dictated by the plasmon mode, $\Pi(k,\omega_n) \approx -nk^2/m\omega_n^2$, see thorough discussion in Ref. [16]. Moreover, we find that this unphysical behavior can be traced back to the numerical observation that the GW approximation does not respect the dynamic particle number conservation law, which implies that at zero momentum $\Pi(k=0,\omega_n) \propto \delta_{n,0}$, or, identically, $\Pi(k=0,\tau) = \text{const}$. Indeed, for an arbitrary interaction potential Π is related to the density-density correlation function, χ, via $\Pi = -\chi/(1 - V\chi)$, while $\chi(k=0,\tau) = \langle \hat{N}(0)\hat{N}(\tau)\rangle \equiv \text{const}$ because the total number of particles $\hat{N}$ commutes with the Hamiltonian. As a result, at zero momentum the density-density response is purely static, $\chi(k=0,\omega_n \neq 0) \equiv 0$, see Ref. [16]. Instead, within the GW approximation, one finds that $\Pi(k=0,\omega_n)$ has significant amplitudes at finite frequencies, and, correspondingly, $\Pi(k,\omega_n \neq 0)$ is not approaching zero when $k \to 0$. This also causes significant problems for the proper technical implementation of the GW approach in Coulomb systems because $(4\pi e^2/k^2)\Pi(k,\omega_n)$ tends to diverge at small momenta and forces one to consider extremely large frequencies in the calculation of the screened interaction W.

Since all problems originate from the violation of the dynamic particle conservation law, we propose a simple strategy to enforce the physical behavior of $\Pi(k,\omega_n)$. All one has to do is to perform a transformation

$$\Pi(k,\omega_n) \to \Pi(k,\omega_n) - \Pi(0,\omega_n) + \Pi(0,0)\delta_{n,0}\ , \qquad (5)$$

before calculating the dielectric response from the GW solution. In other words, one has to subtract the spurious frequency dependence at $k = 0$. Note that this transformation is compatible with higher-order diagrammatics and we suggest that it should be implemented within the fully self-consistent skeleton schemes which involve the self-consistent determination of Π. Indeed, in the large-order expansion limit the correction term is supposed to vanish when $\Pi(k=0,\omega_n)$ converges to the correct physical behavior $\propto \delta_{n,0}$.

In Figs. 2 and 3 we show how our protocol works in practice by considering the case of $r_s = 1$ at low temperature $T/\epsilon_F = 0.02$ and small momentum $k/k_F = 0.1$. First, we performed analytic continuation of the imaginary frequency data for $\epsilon(k,\omega_n)$ using a hybrid of stochastic optimization [17, 18] and consistent constraints [19, 20] methods to get the imaginary part $\epsilon''(k,\omega)$. Next, the real part $\epsilon'(k,\omega)$ is obtained from the Kramers-Kronig relation. The improvement in terms of eliminating the unphysical behavior is dramatic. After the transformation, the high-frequency tail of $\epsilon''(k,\omega)$ gets suppressed by nearly two orders of magnitude. As a result, the real part of the dielectric function now has its zero at $\omega_p^{(GW)} \approx 0.89(1)\omega_p$ and is approaching unity from below at $\omega \gg \epsilon_F$. [In order to have $\omega_p^{(GW)}$ to coincide with ω_p precisely, one would need to divide Π by Z^2, mimicking the effect of vertex corrections.] Everything about the original GW data at frequencies $\omega > kv_F$ is completely unsatisfactory.

Ground-state properties. Since we need G_σ anyway in order to calculate Π via Eq. (2) we can easily extract

TABLE I. Minus the ground-state exchange-correlation energy per particle $-E_{XC}$ (in Hartree), the quasi-particle residue Z, and the effective mass renormalization m_*/m at the Fermi level for the unpolarized 3D homogeneous electron gas. All these quantities are obtained by using the *standard* GW approach, i.e. solving the set of Eqs. (1)-(4).

r_s	1	2	4	5	10
$-E_{XC}$	0.5267(2)	0.2789(1)	0.1488(1)	0.1216(1)	0.06498(2)
Z	0.899(1)	0.842(1)	0.769(2)	0.743(2)	0.658(2)
m_*/m	0.944(2)	0.931(2)	0.913(2)	0.906(2)	0.875(2)

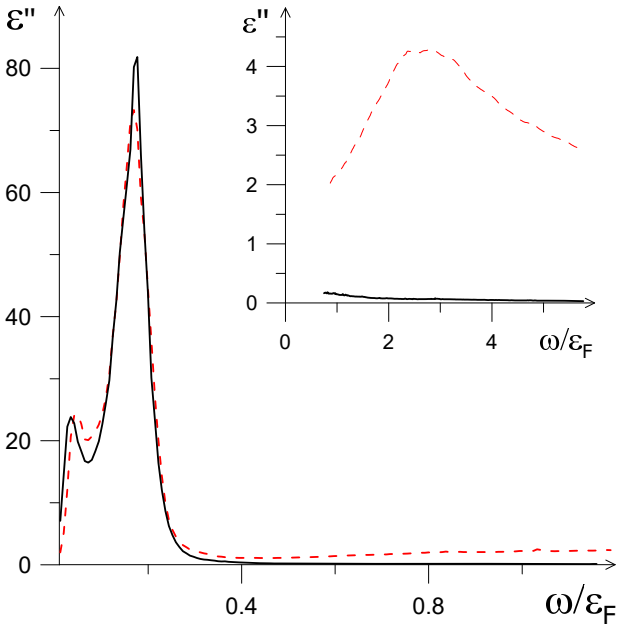

FIG. 2. Color online: Imaginary part of the dielectric function within the GW approximation at $r_s = 1$, $k/k_F = 0.1$, and $T/\epsilon_F = 0.02$. Red dashed curve is the original GW result, and the solid black line is the corrected GW spectrum. The crucial difference at frequencies $\omega > kv_F$ is clearly seen in the inset.

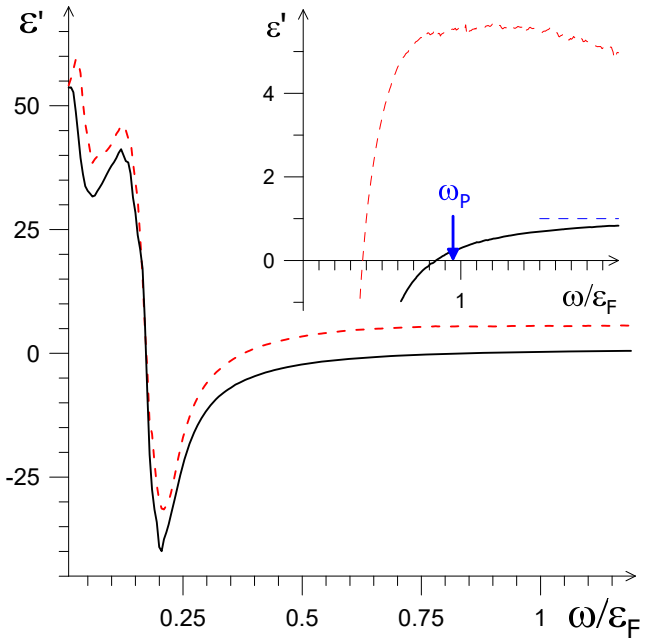

FIG. 3. Color online: Real part of the dielectric function within the GW approximation at $r_s = 1$, $k/k_F = 0.1$, and $T/\epsilon_F = 0.02$. The original GW result (red dashed curve) completely misses the plasmon zero, and predicts unrealistically large response at frequencies above ϵ_F. The corrected result (solid black line) crosses zero within 10% of ω_p and saturates to unity at $\omega > \epsilon_F$.

Fermi-liquid properties from it and check them against known results. The GW technique, self-consistently solving the above set of Eqs. (1)-(4), was implemented in the past for jellium at zero temperature in Refs. [8, 13, 14] and at finite temperature in Ref. [15]. It was concluded [13, 14] that the method produces ground-state energies that agree with diffusion Monte Carlo results [21] at the sub-percent level (see also Table in the Supplementary material). Apparently, this conclusion was based on data containing some systematic bias. We find that our exchange-correlation energies differ from those of Refs. [8, 13, 14] well outside the error bounds and by an amount bigger than the difference between the GW and various other approximations, for instance $GW^{(0)}$. To ensure correctness of our results, we developed two absolutely independent codes that did not share a single common idea about grids and cutoffs for storing and processing the data, Fourier transforms, and energy evaluation. Moreover, the second code was implemented for the Yukawa potential and final results were recovered by extrapolating the Yukawa screening wavevector to zero. See Supplemental Material at [URL will be inserted by publisher] for details about these two different implementations. Our data for the ground-state energy is found to be in agreement only with the one plotted in Ref. [15]. Ultimately, four [22] independently developed finite-T codes were compared and found to be in agreement with each other within the error bounds reported in Table I.

For benchmark purposes we report here the ground-state exchange-correlation energy, the quasi-particle Z-factor, and the effective mass renormalization in Table I. Error bounds were estimated from variations induced by changing momentum-time grids, cutoffs, and extrapolation procedures to the zero-temperature limit. All re-

sults in the table were obtained for the *standard* GW formulation; i.e., the transformation procedure (5) was *not* applied when solving Eqs. (1)-(4).

While applying the transformation (5) to the GW solution vastly improves the two-body spectral properties, it is natural to ask what impact it has on the Fermi-liquid properties when applied at each iteration of the self-consistent scheme; we abbreviate the corresponding scheme as GW(Π). The exchange-correlation energy, Z-factor and effective mass obtained in that way are given in Table II. The relative change in exchange-correlation energy ranges from about 1% for $r_s = 1$ up to almost 10% for $r_s = 10$. Given that the relative difference in exchange-correlation energy calculated with the standard GW approach and with the diffusion Monte Carlo method [21] is a few percent, we conclude that the GW(Π) scheme has the same quality in terms of the thermodynamic properties while it improves the two-body spectral function by orders of magnitude.

Conclusions. We have proposed a simple strategy to drastically improve key properties of the two-particle correlation functions within the GW approximation and applied it to the jellium model. The strategy is designed to cure unphysical behavior of the polarization function Π that originates from the violation of the dynamic particle conservation law. The very same trick can be applied to other models and materials science systems, and can be used in the Diagrammatic Monte Carlo approach that considers higher-order vertex corrections (which should correct the unphysical behavior of Π). We also report benchmark values of key Fermi liquid parameters for jellium within the *standard* GW approximation and the *modified* GW(Π) version.

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[22] After this work was completed our values for the ground-state energy were confirmed by yet another (fourth) implementation by L. Pollet.

TABLE II. Minus the ground-state exchange-correlation energy per particle $-E_{XC}$ (in Hartree), the quasi-particle residue Z, and the effective mass renormalization m_*/m at the Fermi level for the unpolarized 3D homogeneous electron gas. These are obtained by using the *modified* GW, or GW(Π), approach, i.e. by solving the set of Eqs. (1)-(4) self-consistently and at the same time applying the transformation Eq. (5) to Π at each iteration of the self-consistent scheme.

r_s	1	2	4	5	10
$-E_{XC}$	0.5205(2)	0.2716(2)	0.1413(1)	0.1143(1)	0.05873(2)
Z	0.880(3)	0.808(2)	0.722(2)	0.692(3)	0.605(4)
m_*/m	0.934(3)	0.900(2)	0.834(2)	0.803(2)	0.673(4)