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A COUPLED-CLUSTER ANALYSIS OF ONE-COMPONENT COULOMB PLASMAS

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The coupled-cluster formulation of quantum many-body theory has been discussed in some detail by Lührmann and the present author [1], where attention was focussed on the problem of ground-state correlations in the one-component electron plasma (or "electron gas" or "jellium") and the associated problems due to the long-range interactions and correlations. This initial work was restricted to the high-density limit and we showed then how to solve the problem exactly within the coupled-cluster formalism (CCF) in both the random-phase approximation (RPA) and the Tamm-Dancoff approximation (TDA) -- both of which are mathematically non-trivial in the CCF. This earlier CCF analysis has now been extended [2] into both the low-density regime and to intermediate densities appropriate to real metals, and it is this work that now forms the main topic for discussion.

As is by now well known, the CCF exactly decomposes the N-body system into a set of mutually interacting n-body subsystems ($n = 1, \cdots, N$) by use of the familiar exp(S) Ansatz acting with reference to some quite arbitrary model (or reference or non-interacting) N-body state, which we shall consistently take to be the usual filled Fermi sea (or Slater determinant of plane-wave states) appropriate to an unpolarized, spin-$\frac{1}{2}$, homogeneous system. The exp(S) Ansatz is then really just a convenient rewriting of the usual linked-cluster theorem for the ground-state (g.s.) wavefunction. The resulting subsystems are thus described by a set of correlation operators S_n, whose matrix elements give the amplitudes for exciting n particle-hole pairs from the model state. The exact N-body g.s. Schrodinger equation may then formally be decomposed into a coupled set of N nonlinear ("linked") microscopic equations for these matrix elements, or subsystem amplitudes, in which all macroscopic terms (i.e., those proportional to N^{ν} for $\nu \geq 1$) are absent.

The general structure of this set of equations is that the n^{th} equation in the hierarchy for S_n is coupled, in the general case where the fundamental hamiltonian contains up to j-body interactions, to all higher amplitudes S_{n+i} with $1 \leq i \leq j$ as well as to all lower S_{ℓ} with $\ell < n$. In the present case we are concerned only with the $j = 2$ Coulomb interaction. Clearly, in order to be of practical use, this hierarchy of equations (which is otherwise an exact decomposition for the N-body g.s.) must be truncated, and the so-called "natural" or SUBn approximation scheme, with which we shall largely be concerned henceforth, does this by setting each S_{ℓ} to zero for $\ell > n$. In the case of translationally-invariant systems, momentum conservation implies $S_1 \equiv 0$, and we shall thus initially focus attention on the SUB2 approximation. Working in the natural single-particle momentum-eigenstate (i.e. plane-wave) basis appropriate for an infinite, homogeneous system, the SUB2 equations are nonlinear integral equations in three three-momenta for the (antisymmetrized) matrix elements,

$$S_2(\vec{k}_1, \vec{k}_2; \vec{q}) \equiv \langle \vec{k}_1 + \vec{q}, \vec{k}_2 - \vec{q} | S_2 | \vec{k}_1, \vec{k}_2 \rangle_A$$

where the "hole" states are labelled by momenta $\vec{k}_1$ and $\vec{k}_2$ inside the Fermi sphere, and where spin labels have been suppressed for ease of notation.

It is worth stressing again that the basic SUB2 approximation is exact apart from neglecting interactions with higher-order subsystems. Otherwise, all two-body effects are included, and it is not therefore surprising that the equations are complex. As drastic sub-approximations to itself, the SUB2 approximation wholly contains such other familiar approximations as RPA and TDA; the Bethe-Goldstone equation which sums the two-particle "ladder" series of diagrams, and more generally the whole of (lowest order) Brueckner theory; and the Galitskii approximation which represents the ladder approximation to the Bethe-Salpeter equation.

In the high-density limit (i.e., the weak-coupling limit $r_s \to 0$, in terms of the dimensionless coupling parameter r_s, which is the average interparticle spacing in units of the Bohr radius), the RPA gives the leading contribution to the correlation energy ε_c (i.e. the g.s. energy per particle relative to the energy of the uncorrelated model state, which is also just the energy in the (Hartree-) Fock approximation, with all energies expressed in Rydberg units). The nonlinear (and highly non-trivial!) RPA integral equation for S_2 was solved exactly and in some detail in Ref. [1].

In the intermediate-coupling regime ($1 \lesssim r_s \lesssim 5$) appropriate to metallic densities, it is certainly not expected that the RPA will remain a good approximation. Thus, quite apart from ignoring (i) even the simple exchange terms needed to antisymmetrize the RPA itself, we have further neglected, even in SUB2 approximation: (ii) the combinations of two-particle and two-hole ladder terms, some at least of which are vital for correct short-range behaviour; (iii) the self-energy correction terms which self-consistently generate both the particle potential and, more importantly, the hole potential; (iv) classes of particle-hole ladder interaction terms; and (v) higher exchange terms which are necessary to preserve the overall antisymmetry. In this last context it is also worth noting that the fermion statistics are exactly obeyed in each complete SUBn truncation, although further sub-approximations such as the RPA may of course violate them. In order now to proceed systematically beyond the RPA by including these extra SUB2 effects, we have developed a further necessary "state-averaging" approximation in order to make the resulting SUB2 equations amenable to numerical solution. This approximation, which is motivated by analogy to the much simpler equations for the corresponding many-boson system, takes the basic form of averaging over the interior of the Fermi sea the hole momenta $\vec{k}_1$ and $\vec{k}_2$ in $S_2(\vec{k}_1, \vec{k}_2; \vec{q})$, but at the same time preserving the Pauli principle by requiring that the particle momenta $(\vec{k}_1 + \vec{q})$ and $(\vec{k}_2 - \vec{q})$ lie outside the Fermi sea. In this way the exact $S_2(\vec{k}_1, \vec{k}_2, \vec{q})$ is replaced by an averaged $\langle S_2(q) \rangle$,

$$S_2(\vec{k}_1, \vec{k}_2; \vec{q}) \to \langle S_2(q) \rangle \, \theta(\vec{k}_1, \vec{q}) \, \theta(-\vec{k}_2, \vec{q}) \quad ,$$

$$\theta(\vec{k}, \vec{q}) \equiv \theta(k_F - k) \, \theta(|\vec{k} + \vec{q}| - k_F)$$

where $\theta(x)$ is the usual unit-step function defined to be one (zero) for x greater (less) than zero, and k_F is the Fermi momentum. The resulting CCF equation must now still be suitably averaged over $\vec{k}_1$ and $\vec{k}_2$, and although this latter step is clearly not unique, we may turn this to our advantage by

making a suitable choice on physically-motivated grounds. Furthermore, since we know exact results for S_2 in at least one limit, namely the RPA and TDA results for $r_s \to 0$, the errors induced by the state-averaging procedures may be checked and controlled in this limit at least.

As a simple illustration of the above procedure, let us imagine putting it into effect for the RPA which in our CCF language comprises an equation for S_2 which involves only the one-body bare kinetic energy (KE) and the RPA terms (and see Ref. [1] for further details). After the replacement $S_2 \to \langle S_2 \rangle$ as indicated above, the only dependence on hole-state momenta is in the KE term which takes the very familiar form of being proportional to

$$[|\vec{k}_1 + \vec{q}|^2 + |\vec{k}_2 - \vec{q}|^2 - k_1^2 - k_2^2] \langle S_2(q) \rangle \equiv e \langle S_2(q) \rangle \quad .$$

As two obvious candidates for the final averaging one could now imagine either (i) replacing $e \to \langle e \rangle$; or (ii) the intuitively and physically more appealing procedure of first dividing throughout by e, and then replacing $e^{-1} \to \langle e^{-1} \rangle$, i.e. averaging the two-particle bare propagator or "energy denominator". We show that the former procedure leads precisely to the "mean spherical approximation" discussed by Zabolitzky [3] in this context via his state-independent variational treatment of the electron gas in the Fermi hypernetted chain (FHNC) formalism, in this same $r_s \to 0$ limit. Whereas this approximation gives $\varepsilon_c \to 0.0570 \ln r_s$ as $r_s \to 0$, which is 8.4% in error by comparison with the exact result, the latter $\langle e^{-1} \rangle$-averaging scheme is in fact exact in this limit (giving $\varepsilon_c \to 0.0622 \ln r_s$), and over the entire range $1 \leq r_s \leq 5$ gives results to within 2% of the exact RPA results. Furthermore, there is no reason to expect markedly worse accuracy for any of the other terms in the Fermi SUB2 equations.

Our most complete results for metallic densities include the completely integrated and self-consistent effects of the terms which by themselves generate: (i) RPA and its exact long-range screening effects; (ii) the extra exchange effects to preserve antisymmetry in the RPA; (iii) the self-consistent particle-particle ladders (LAD) that describe two-electron scattering within the many-electron system, and which describe the exact short-range limiting behaviour; (iv) a class of particle-hole ladder diagrams; and (v) the self-consistent hole-potential. Particular attention is paid to the important effects caused by the interference at intermediate separations of the long-range RPA and short-range LAD effects; and this in fact even leads us to go partially beyond SUB2 approximation to include a much broader class of generalized ladder terms by incorporating part of the coupling to the three- and four-body subsystem amplitudes S_3 and S_4 respectively, and which are hence no longer set to zero. The outcome of this partial inclusion of the coupling terms to higher subsystems is that the bare Coulomb potential which appears in the other SUB2 terms is replaced by a self-consistent G-matrix (obtained from the full S_2 solution itself). Finally we also give arguments that all other SUB2 terms either approximately cancel among themselves, or are negligibly small at metallic densities.

The results of our coupled cluster calculation so described are shown in Table I in the column headed CC, and are there compared with RPA results and results from several other recent calculations. The electron gas is probably the most well-studied of all quantum many-body problems, and hence a vast number of calculations on it exist. We restrict comparison therefore to some few representative alternative approaches. Since the RPA was developed by Bohm and Pines [4], much work has been invested in going beyond it to incorporate the effects due to short-range correlation and exchange

Table I. The correlation energy per particle, ε_c, in Rydbergs for the unpolarized electron gas at various densities.

r_s	$-\varepsilon_c$						
	RPA	RPA+ RPAEX(1)	CC	GFMC	AP	FHNC	VS
1	0.1576	0.1182	0.123	0.121	0.117	0.1141	0.130
2	0.1236	0.0885	0.0917	0.0902	0.086	0.0859	0.098
3	0.1055	0.0733	0.0751	(0.074)	0.069	0.0710	0.081
4	0.0936	0.0637	0.0644	(0.064)	0.058	0.0612	0.070
5	0.0849	0.0568	0.0568	0.0563	0.051	0.0541	0.062

which are so important in the metallic density range. For example, Singwi and his co-workers (see e.g. Refs. [5,6] and other references cited therein) developed a phenomenology based on the dielectric function which attempted to include the effects of the "Coulomb hole" created around each electron by exchange effects and the correlations due to two-particle interactions. The results of Vashishta and Singwi [6], which represent perhaps the culmination of this approach, are shown for comparison in Table I in the column labelled VS. Within the framework of perturbation theory similar considerations have led many authors to improve upon RPA by including also the electron-electron ladder diagrams. Typical of this approach is that of Lowy and Brown [7], who obtained very similar results for the correlation energy to Singwi et al. [5], although their methods are quite different. The column headed RPA+RPAEX(1) in Table I, which is included for purposes of later comparison, represents the approximation of including all RPA ring diagrams plus all such rings where only a single (first-order) exchange (i.e., between only one pair of electrons) is allowed.

More recently two new interesting approaches to this old problem have appeared. The first of these is the FHNC method. This is a variational method based on a many-body wavefunction of Jastrow type, in which the expansion for the two-body radial distribution function is truncated in the so-called Fermi hypernetted chain (FHNC) approximation (thereby losing the variational bound on the energy), and whence a self-consistent scheme emerges to calculate the distribution function. The FHNC results of Zabolitzky [3] are shown in Table I as being representative of the best of the currently available variational results for this system.

The second recent study is that due to Arponen and Pajanne [8], who employ a boson formulation of the many-fermion problem in which the basic idea is to find suitable boson images for the fermion particle-hole pair operators, which satisfy the same exact commutation relations as their fermion-pair counterparts. A further canonical transformation is then made to the Sawada bosons which are just the elementary RPA excitations. The resulting boson hamiltonian then contains the RPA as the lowest order (non-interacting) piece, plus the further terms which represent interactions among the Sawada bosons and which go beyond the RPA. The full interacting boson hamiltonian is then treated by Arponen and Pajanne by the bosonic exp(S) or CCF method, and they also employ the analogous SUB2 approximation. Their results are shown in Table I in the column headed AP.

Finally, all of these calculations may be compared with the essentially exact benchmark results of Ceperley and Alder [9] in the column headed GFMC, which derive from the very lengthy computer runs on stochastic simulations of the many-body Schrödinger equation employing Green function Monte Carlo (GFMC) techniques. Since these results were not given for $r_s = 3, 4$, the corresponding values in parentheses in this column have been interpolated from the computed values shown by Vosko et al. [10], using a Padé approximant technique. It is clear by comparison that our fully microscopic and non-phenomenological results over the entire metallic density range are at least as good as any of the best of the alternative approaches. It is also interesting to note that inclusion of just first-order exchange terms beyond RPA gives remarkably good results for the correlation energy. We believe this result is essentially fortuitous, since for quantities other than the fully-integrated energy the RPA+RPAEX(1) approximation is surely not expected to give such good agreement. However what this result clearly shows is that the sum of all the higher-order diagrams, which are undoubtedly individually very important for a proper description of the electron liquid at these densities, is close to zero. It is undoubtedly for just this reason that the electron gas has proved such a difficult many-body problem.

Turning finally to the very interesting strong-coupling limit, $r_s \to \infty$, one may not at first sight expect the SUB2 approximation to give a reasonable approximation since one imagines that n-body clusters with $n \gg 2$ are of importance here. Indeed we expect the system to undergo a phase transition to a Wigner solid, and such a crystal phase is archetypal of a situation where N-body correlations dominate. However, in SUB2 approximation we find that $\varepsilon \to Ar_s^{-1} + Br_s^{-3/2} + O(r_s^{-2})$ as $r_s \to \infty$. (Strictly we prove this result only for the one-component boson Coulomb plasma, but we note that the exact effects of quantum statistics becomes exponentially small in this limit.) The KE term contributes only to the constant B in leading order, and whereas each of the three remaining terms (in boson SUB2 approximation) is necessary for a quantitative evaluation of the constant A, they play quite different physical roles. Thus whereas the RPA terms continue to be vital for the correct analytic behaviour by providing the correct long-range Coulomb screening, the condensate potential (which for boson systems is the analogue of the hole potential for fermions) now plays a similarly crucial role in the short-range limit. Whereas the inclusion of the only remaining term (namely the two-particle ladder term) quantitatively changes the constants A and B it may safely be omitted without changing the analytic form of the low-density energy expansion. The virial theorem may also be used to show that in this low-density expansion the leading term is purely potential energy, whereas the much more interesting second term is half each kinetic and potential energy -- a result strongly reminiscent of the simple harmonic motion (SHM) expected of the zero-point motion of a solid. Indeed, Wigner first pointed out that in this limit where the potential energy dominates, the energy is minimized by the particles crystallizing, and this leads to an electrostatic energy $\sim r_s^{-1}$. Whereas in a fluid phase the particles are free to occupy the whole volume and hence by the uncertainty principle have a KE $\sim r_s^{-2}$, the particles in the Wigner solid are constrained to oscillate about fixed lattice sites and hence to have a greater KE, which by SHM considerations is readily seen to vary as $r_s^{-3/2}$. Thus our SUB2 g.s. energy has the correct analytic form of a solid, even though the coefficients A and B are not very close to the true g.s. results for a b.c.c. crystal. It is however important to realize that even the lowest SUB2 approximation in the CCF gives a low-density energy which cannot possibly represent a fluid phase since the particles are not free to occupy the whole volume.

It is clear therefore that our everywhere translationally-invariant g.s. CCF approach well describes both fluid and solid phases, all the way from the exact high-density RPA plasma limit, through the intermediate-coupling range of

metallic densities, to the (strong-coupling) low-density crystalline solid phase which is now approximated as an amorphous, glassy solid phase. Although one might be surprised that our fluid-like SUB2 wavefunction (built from the non-interacting model state) is capable of describing an (amorphous) solid phase, it is clear that strong many-body correlations are built into it. On the other hand it is clear that in this low-density limit the third- and higher-order correlations obtained from the (so-far) neglected coupling terms to the amplitudes S_n with $n > 2$, are still very strong, and need to be incorporated for a better quantitative description. Nevertheless, the SUB2 description of the one-component Coulomb plasmas is seen to be remarkably successful over the entire density range, and now gives what is perhaps the best available microscopic description of the g.s. of this most well-studied of all quantum many-body systems.

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