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One-component charged systems as a limiting case of quantal two-component mixtures—a bifurcation procedure for a many-body system

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Abstract. The standard model problem of a three-dimensional fully interacting electron gas in a uniform compensating background is shown to result from a procedure starting at the operator level with a fully quantum mechanical *two-component* system of electrons and countercharge (e.g. nuclei), and following a dual limiting process involving the division of charge and the scaling of mass. The bifurcation method provides insight into other aspects of the corresponding many-body problem, two examples of which are given.

1. Introduction

In the non-relativistic limit the Hamiltonian for an element of atomic number Z_a consisting of N nuclei ($\alpha = \mathrm{n}$) and $Z_a N$ electrons ($\alpha = \mathrm{e}$) can be written

$$\hat{H} = \sum_{\alpha}\left(\hat{T}_\alpha + \frac{1}{2}\sum_{\alpha'}\int \mathrm{d}\boldsymbol{r}\int \mathrm{d}\boldsymbol{r}' Z_\alpha Z_{\alpha'} v_c(\boldsymbol{r}-\boldsymbol{r}')\hat{\rho}^{(2)}_{\alpha\alpha'}(\boldsymbol{r},\boldsymbol{r}')\right) \tag{1}$$

where $Z_\alpha = Z_a$ for $\alpha = \mathrm{n}$, and $Z_\alpha = -1$ for $\alpha = \mathrm{e}$. In 1 $v_c(\boldsymbol{r}-\boldsymbol{r}') = e^2/|\boldsymbol{r}-\boldsymbol{r}'|$ is the basic Coulomb interaction; $\hat{T}_\alpha = \sum_{i=1}^{N_\alpha}(-\hbar^2/2m_\alpha)\nabla^2_{\alpha i}$ are the kinetic contributions. The quantity $\hat{\rho}^{(2)}_{\alpha\alpha'}$ is the the two-particle density operator defined by

$$\hat{\rho}^{(2)}_{\alpha\alpha'}(\boldsymbol{r},\boldsymbol{r}') = \hat{\rho}^{(1)}_\alpha(\boldsymbol{r})\hat{\rho}^{(1)}_{\alpha'}(\boldsymbol{r}') - \delta_{\alpha\alpha'}\delta(\boldsymbol{r}-\boldsymbol{r}')\hat{\rho}^{(1)}_\alpha(\boldsymbol{r}) \tag{2}$$

where

$$\hat{\rho}^{(1)}_\alpha(\boldsymbol{r}) = \sum_{i=1}^{N_\alpha}\delta(\boldsymbol{r}-\boldsymbol{r}_{i\alpha}) \tag{3}$$

is the standard one-particle density operator for instantaneous coordinates $\{\boldsymbol{r}_{i\alpha}\}$. The simplicity and symmetry of (1) leads to exact scaling results for the ground state energy and the structure functions [1] which will be used in the following. According to the choice of external conditions (the choice of volume V containing a canonical, neutral, ensemble) the states of (1) usually divide into two distinct classes which in one-electron language we would describe as localized-core orbitals (close to the atomic equivalents), and itinerant valence states leading for crystalline arrangements to Bloch levels and a corresponding band structure. A first approximation to the latter is often taken to be the one-electron

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structure taken from a many-body problem in which the core structure, the combination of nuclear charge and core-orbital charge, is replaced by a uniform background, and all further consequences of the fermionic character of the localized electrons eliminated. This leads to the standard many-body problem for a fixed number of interacting electrons in a neutralizing background. We show here for a fully two-component arrangement how this problem can be arrived at from the level of the Hamiltonian (1) by a systematic procedure involving the division of charge and the scaling of mass.

2. A bifurcation model

Setting $Z_a = Z$ in what follows, start again with a neutral system of NZ negative point fermions (electrons) each of mass m_e and charge $-e$, and N positive point counterions (also assumed to be fermions) each of mass M and charge Ze ($e > 0$). The fully dynamical problem for this system is described by the two-component Hamiltonian (1) with Coulomb interactions. Proceed now with the following formal device, which is justified by the universal applicability of quantum mechanics to physical systems with widely differing masses and charges. First, split every counterion into two point particles each of charge $Ze/2$ but at the same time *increase* the mass of each new particle to σM, the coefficient σ being determined later ($\sigma > 1$, as will be seen in equation (9) below). We may take the counterions as of either fermionic or bosonic character, although it is important that the symmetry class is preserved under division. If we iterate this procedure to the νth step, the transformation of the counterion subsystem can be described by

$$(N, M, Ze) \rightarrow (2^\nu N, \sigma^\nu M, Ze/2^\nu).$$

Now the NZ electrons together with the $2^\nu N$ positive particles consists of a new but still neutral canonical system described by the Hamiltonian

$$\hat{H} = \sum_{i=1}^{NZ} \frac{p_{i,e}^2}{2m_e} + \frac{1}{2}\sum_i \sum_{j\neq i} \frac{e^2}{|\boldsymbol{r}_{i,e} - \boldsymbol{r}_{j,e}|} + \sum_{i=1}^{2^\nu N} \frac{p_{i,p}^2}{2\sigma^\nu M} + \frac{1}{2}\sum_i^{2^\nu N} \sum_{j\neq i}^{2^\nu N} \frac{(Ze/2^\nu)^2}{|\boldsymbol{r}_{i,p} - \boldsymbol{r}_{j,p}|} - \sum_{i=1}^{NZ}\sum_{j=1}^{2^\nu N} \frac{Ze^2/2^\nu}{|\boldsymbol{r}_{i,e} - \boldsymbol{r}_{j,p}|}. \tag{4}$$

It clearly continues to describe a fully dynamical two-component system, and it will also therefore be constrained by identical formal scaling relations $([NZ, m_e, -e], [2^\nu N, \sigma^\nu M, Ze/2^\nu])$. We will now show that in the limit $\nu \rightarrow \infty$ we recover just the intuitively anticipated problem of an interacting electron gas in a uniform *static* background (i.e., the first two terms of (4) plus background terms), provided only that we choose σ appropriately (as noted according to equation (9) below). To do this we observe the following:

From the cusp condition [2, 3, 4] (but now applied for two positive particles of charge $Ze/2^\nu$ and mass $\sigma^\nu M$ each) we have

$$\left.\frac{\partial \rho_{\alpha\alpha}(r)}{\partial r}\right|_{r=0} = \frac{2\rho_{\alpha\alpha}(r=0)}{a^*} \tag{5}$$

where $\rho_{\alpha\alpha}(r)$ is the sphericalized average of the two-particle density operator (equation (2)) for the positive particles ($\alpha = \mathrm{n}$). In (5) $a^* = \frac{\hbar^2}{(\sigma^\nu M/2)(Ze/2^\nu)^2}$ is the effective Bohr radius for these particles. Accordingly

$$\left.\frac{\partial \rho_{nn}(r)}{\partial r}\right|_{r=0} \propto \left(\frac{\sigma}{4}\right)^\nu$$

and it is clear therefore that if we choose $\sigma < 4$ we must have

$$\lim_{\nu\to\infty} \left.\frac{\partial \rho_{nn}(r)}{\partial r}\right|_{r=0} = 0.$$

This means that in the $\nu \to \infty$ limit the cusps rigorously vanish, and in any physical states of interest the density of the positive subsystem of the new particles must be uniform. An immediate consequence is that no fluctuations for the positive subsystem exist in this limit, and for this reason we can now replace operators with their averages. In particular, since the one-particle density operator for the positive particles in the νth step (equation (3)) has an average

$$\langle \hat{\rho}_n^{(1)}(\boldsymbol{r})\rangle = 2^\nu \rho_n^{(1)}(\boldsymbol{r})$$

we have the result that in the limit $\nu \to \infty$ we can replace the operator

$$\lim_{\nu\to\infty} \frac{1}{2^\nu}\hat{\rho}_n^{(1)}(\boldsymbol{r})$$

with its average N/V. This is constant because as just shown above the one-particle density of the positive subsystem in this limit will be uniform. Consider, then, the last two terms of (4). First

$$\frac{1}{2}\sum_{i}^{2^\nu N}\sum_{j\neq i}^{2^\nu N}\frac{(Ze/2^\nu)^2}{|\boldsymbol{r}_{i,p}-\boldsymbol{r}_{j,p}|}$$

can be written as

$$\lim_{\lambda\to 0}\frac{1}{2}\left(\frac{Ze}{2^\nu}\right)^2\int\int \mathrm{d}\boldsymbol{r}\mathrm{d}\boldsymbol{r}'\,\hat{\rho}_{nn}^{(2)}(\boldsymbol{r},\boldsymbol{r}')\frac{\mathrm{e}^{-\lambda|\boldsymbol{r}-\boldsymbol{r}'|}}{|\boldsymbol{r}-\boldsymbol{r}'|}$$

where the two- and one-particle density operators appearing are defined by (2) and (3) and, according to the above result, in the limit $\nu \to \infty$ this approaches

$$\lim_{\lambda\to 0}\frac{Z^2}{2}e^2\int\int \mathrm{d}\boldsymbol{r}\mathrm{d}\boldsymbol{r}'\frac{\frac{N}{V}\frac{N}{V}\mathrm{e}^{-\lambda|\boldsymbol{r}-\boldsymbol{r}\ '|}}{|\boldsymbol{r}-\vec{\boldsymbol{r}}\ '|} = \lim_{\lambda\to 0}\frac{1}{2}Z^2e^2\frac{N^2}{V}\frac{4\pi}{\lambda^2}. \tag{6}$$

Similarly the term

$$-\sum_{i=1}^{NZ}\sum_{j=1}^{2^\nu N}\frac{Ze^2/2^\nu}{|\boldsymbol{r}_{i,e}-\boldsymbol{r}_{j,p}|}$$

can be written as

$$\lim_{\lambda\to 0}\left[-\frac{Ze^2}{2^\nu}\int\int \mathrm{d}\boldsymbol{r}\mathrm{d}\boldsymbol{r}'\,\hat{\rho}_e^{(1)}(\boldsymbol{r})\hat{\rho}_n^{(1)}(\boldsymbol{r}')\frac{\mathrm{e}^{-\lambda|\boldsymbol{r}-\boldsymbol{r}'|}}{|\boldsymbol{r}-\boldsymbol{r}'|}\right]$$

and in the limit $\nu \to \infty$ it becomes

$$\lim_{\lambda\to 0}\left[-Ze^2\sum_{i=1}^{NZ}\int \mathrm{d}^3r\frac{N}{V}\frac{\mathrm{e}^{-\lambda|\boldsymbol{r}-\boldsymbol{r}_i|}}{|\boldsymbol{r}-\boldsymbol{r}_i|}\right] = \lim_{\lambda\to 0}\left[-Z^2e^2\frac{N^2}{V}\frac{4\pi}{\lambda^2}\right]. \tag{7}$$

Here λ is the standard convergence parameter for long-ranged interacting systems that is ultimately taken to zero, but always in such a way as to keep $\lambda^{-1} << V^{1/3}$ in the approach to the thermodynamic limit.

Lastly we have to deal with the kinetic energy term (the third term in (4)). Because, as was shown above, for the limit $\nu \to \infty$ the positive particle subsystem is in a state of uniform density and also infinitely dense, and also because each particle has asymptotically a vanishing value of charge, this subsystem in this limit can be considered as an ideal system.

We are therefore confronted with an ideal Fermi system of density $\lim_{\nu\to\infty}(2^\nu N)/V$ of particles, each with mass $\lim_{\nu\to\infty}\sigma^\nu M$. In this case the third term of (4) will contribute a kinetic energy

$$T_p = \frac{m_e}{M\sigma^\nu}\frac{2.21}{r_s^2} \quad \text{Ryd} \tag{8}$$

per particle. However, $V/(2^\nu N) = \frac{4}{3}\pi r_s^3 a_0^3$ so that $r_s^2 \propto 1/(2^{2\nu/3})$ or $r_s^2\sigma^\nu \propto (\sigma/4^{1/3})^\nu$. It immediately follows that if we choose $\sigma > 4^{1/3}$ we must obtain a vanishing contribution for the kinetic energy per particle in the limit $\nu \to \infty$ for the positive subsystem. (Note also that any residual kinetic term that may result from correlation energy for finite ν is several orders of magnitude smaller than (8) and will also vanish in the limit $\nu \to \infty$.) For a countercharge of boson symmetry, the process of charge division must eventually lead to a non-interacting system and its ground state energy must also approach zero.

We therefore obtain the result that the overall contribution of the positive component in this limit of infinite steps and for any value of σ in the range

$$4^{1/3} < \sigma < 4 \tag{9}$$

will be just the sum of (6) and (7), i.e. $-\frac{1}{2}Z^2e^2(N^2/V)(4\pi/\lambda^2)$, as is the contribution of the positive background in the standard electron gas problem. In the thermodynamic limit this contribution is cancelled by a corresponding electronic one-body term arising as a consequence of charge neutrality, as is well known [5, 6]. Thus after the limit is taken we arrive at the standard one-component Hamiltonian, that can be written as

$$\hat{H} = \hat{T}_e + \frac{1}{2}\int \mathrm{d}\boldsymbol{r}\int \mathrm{d}\boldsymbol{r}' v_c(\boldsymbol{r}-\boldsymbol{r}')\left(\hat{\rho}_{ee}^{(2)}(\boldsymbol{r},\boldsymbol{r}') - 2\rho_0\,\hat{\rho}_e^{(1)}(\boldsymbol{r}) + \rho_0^2\right) \tag{10}$$

where $\rho_0 = ZN/V$.

In the more general case of splitting each nucleus into n (rather than two) nuclei at every step of the bifurcation process the bounds in equation (9) will simply read

$$n^{2/3} < \sigma < n^2 \tag{11}$$

the lower of which is associated with the fermionic character of the positive subsystem. Its existence is traced to the requirement that at each step the mass is sufficient to ensure that the limiting kinetic energy of the nuclei vanishes. The physics of the upper bound is associated with the coulombic character of the interactions, and its existence is traced to the necessity of providing a mass low enough that the limiting density of the system of the nuclei is uniform.

3. Discussion

The above argument shows that by starting with a fully quantum mechanical two-component system a well defined bifurcation procedure applied sequentially converts the formal two-component Hamiltonian to the standard one-component electron gas Hamiltonian in the limit of the procedure and in the thermodynamic limit. We observe that the bifurcation approach may actually be of more general utility for it can provide further insight into other aspects of a many-body charged system. This is demonstrated below in two different contexts: one is related to an exchange driven paired phase of the interacting electron system, and the other to the nontrivial question of the metal–insulator transition in the same problem.

First, the bifurcation procedure, when applied to a moderately dense phase of hydrogen (H_2) molecules (by division of the *protonic* charge and scaling of the protonic mass) is

expected to give in the limit *a paired electron phase* [7] in a uniform background, the pairing being stabilized mostly by exchange. To see this, let us consider the counterion–electron cusp at every step of the bifurcation process, noting that for this problem $Z_a = Z = 1$. We then have

$$\left.\frac{\partial \rho_{\alpha e}(r)}{\partial r}\right|_{r=0} = -\frac{2\rho_{\alpha e}(r=0)}{\tilde{a}} \tag{12}$$

as the analogue of (5), but now with an effective Bohr radius

$$\tilde{a} = \frac{\hbar^2}{m^*(\nu)\left(\frac{Z}{2^\nu}\right)e^2}$$

and with $1/m^*(\nu) = 1/m_e + 1/\sigma^\nu M$. Observe that in the limit $\nu \to \infty$ the cusp vanishes as $1/2^\nu$ for any value of σ. Starting therefore with the usual Heitler–London combination of atomic electronic wavefunctions, with proper cusps, we must obtain vanishing cusps in the limit of the procedure. This is immediately consistent with the use of a Heitler–London combination of Gaussian trial states proposed earlier [7] in a many-body treatment of interacting electrons in a uniform background. This argument therefore gives further support to the possibility of exchange driven pairing at intermediate densities. The ensuing paired-electron phase is then seen as a manifestation of the standard exchange mediated pairing in H_2; it is seen to be a realization of the bifurcation process.

Second, the above procedure can be straightforwardly extended to incorporate in $\hat{H}$ the presence of a gauge field $\boldsymbol{A}$. This has been shown to be an additional important formal device [8] which in this case is capable of addressing the physics of a possible metal–insulator transition in an interacting electron system. The vanishing of the dependence of the ground state energy on $\boldsymbol{A}$ signals the transition to an insulating state, a generalization of an earlier criterion given by Kohn [9]. The simplest method that accounts for the presence of $\boldsymbol{A}$ is the minimal substitution in the kinetic terms of (4) (namely, all the momenta are shifted by $e\boldsymbol{A}/c$). The only change in the above scaling procedure is then in the kinetic energy term of the counterion system. In the limit of repeated bifurcation, equation (8) now becomes

$$T_p = \frac{m_e}{M\sigma^\nu}\left(\frac{2.21}{r_s^2}\ \text{Ryd} + \frac{e^2}{2m_e c^2}\ A^2\right) \tag{13}$$

and it is clear that the new A^2 term will vanish in the $\nu \to \infty$ limit, leaving the presence of $\boldsymbol{A}$ only in the electron kinetic energy term. This turns the problem into one with interacting electrons in a uniform background, but now in the presence of a gauge field. It remains highly nontrivial, but it can be shown to possess a metal–insulator transition that has been estimated earlier (through the use of a random-phase-approximation calculation [8]) to occur at a density corresponding to $r_s = 61.7$. This argument therefore suggests that the metal–insulator transition in this simplest realistic two-component system (the dual fermionic problem originating with electron and protons whose fundamental transitions are still a matter of debate [10]) is mapped by this bifurcation procedure into the well known transition of the interacting electron liquid to the Wigner crystal.

As noted, the bifurcation method we introduce has been applied to fermionic nuclei. We have considered here a three-dimensional system; the extension to two dimensions (or to particles of fractional statistics) may require significant changes [11].

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