



Quantum Coulomb systems: some exact results in the atomic limit

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Abstract

We review a number of exact results concerning the recombined electron–proton gas. The recombination problem can be formulated in precise terms in the atomic limit. In this limit one lets the density and the temperature tend to zero in a coupled way so that the resulting energy–entropy balance favors the formation of certain chemical species. This enables to develop a clear understanding of the dielectric versus conducting behavior in the system. In particular, we give a first principle derivation of the dielectric constant of the dilute atomic gas without presupposing the existence of atoms. The analysis relies on the path integral representation of the Coulomb gas together with Mayer diagrammatic techniques. © 2002 Elsevier Science B.V. All rights reserved.

1. Introduction

Most of the nonrelativistic physics is reputed to be hidden within the N -body Hamiltonian

$$H = \sum_{j=1}^N \frac{|\mathbf{p}_j|^2}{2m_{\alpha_j}} + \sum_{i<j}^N \frac{e_{\alpha_i} e_{\alpha_j}}{|\mathbf{r}_i - \mathbf{r}_j|} \quad (1)$$

describing the Coulomb interaction of N quantum particles (point nuclei and electrons) of species $\alpha = 1, 2, \dots$ with charge e_{α} and mass m_{α} . It is therefore of fundamental interest to derive exact and rigorous results pertaining to this Hamiltonian. A result is *exact* if it can be formally derived without recourse to intermediate approximations or use of models (e.g. asymptotics but without control on the remainder). A result is *rigorous* if its proof meets the professional standards of mathematics. For simplicity, we shall restrict our considerations, in the sequel, to the electron–proton system (e–p)

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in thermal equilibrium at inverse temperature β . We denote by H_{N_e, N_p} the Coulomb Hamiltonian for N_e electrons of charge e and mass m_e and N_p protons of charge $-e$ and mass m_p . We first recall basic rigorous results concerning the thermodynamics of this system (Section 2), and then state some exact results on screening and correlations (Section 3). The used tools (quantum Mayer graphs) are briefly described in Section 4.

2. Rigorous results for thermodynamics

2.1. Stability of matter [1]

There exists a constant $C > 0$ such that

$$H_{N_e, N_p} \geq -C(N_e + N_p), \quad (2)$$

which assures that matter does not implode because of the r^{-1} singularity of the Coulomb potential between charges of opposite sign. In a general multicomponent system, the proof requires that at least one of the particle species (say the electrons) is fermionic. The value of the constant C found in most recent proofs is not too large, but probably not optimal. The stability statement can be formulated in a more refined way. Let E_a be the ground state energy of the hydrogen atom. Then there is a constant B , $0 < B < |E_a|$, such that [3]

$$H_{N_e, N_p} \geq -B(N_e + N_p - 1) \quad \text{for all } (N_e, N_p) \neq (0, 0), (1, 1). \quad (3)$$

The point here is that the constant B can be chosen strictly less than $|E_a|$ for all cases except for the hydrogen atom itself. This is a still unproven, but plausible, conjecture which we shall adopt here as a working hypothesis.

2.2. Existence of the thermodynamic limit [2]

Let $p_\Lambda(\mu_e, \mu_p, \beta)$ be the grand-canonical pressure of the e–p system in a volume Λ , then

$$\lim_{\Lambda \rightarrow \infty} p_\Lambda(\mu_e, \mu_p, \beta) = p(\mu, \beta). \quad (4)$$

The existence of this limit assures that the system does not explode in spite of the long range of the Coulomb potential. Screening is the essential ingredient for the proof. The infinite volume pressure $p(\beta, \mu)$ depends only on the combination $\mu = (\mu_e + \mu_p)/2$ of the chemical potentials μ_e and μ_p of the electrons and protons. This indicates that the system is necessarily locally neutral, namely

$$\rho_e = \rho_p, \quad (5)$$

where ρ_e, ρ_p are the electronic and protonic densities.

2.3. The atomic limit [3–5]

The theorem on the atomic limit is a precise statement about the recombination of electrons and protons into a perfect gas of hydrogen atoms. We introduce the density

of an ideal gas of hydrogen atoms

$$\rho_a^{\text{id}} = \frac{4}{(2\pi\lambda_a^2)^{3/2}} e^{-\beta(E_a - 2\mu)}, \quad \mu = \frac{\mu_e + \mu_p}{2}, \quad (6)$$

where $\lambda_a = \hbar\sqrt{\beta/(m_e + m_p)}$ is the thermal de Broglie length of the atom and the factor 4 accounts for the spin degeneracy. Here all effects of the Coulomb interaction are disregarded except for the binding energy $|E_a|$ of the atom. In the same way, we define the densities of electrons and protons

$$\rho_e^{\text{id}} = \rho_p^{\text{id}} = 2 \left(\frac{\sqrt{m_e m_p}}{2\pi\beta\hbar^2} \right)^{3/2} e^{\beta\mu}. \quad (7)$$

The neutrality is achieved by the special choice $(\mu_e - \mu_p)/2 = (3/4\beta)\ln(m_p/m_e)$ for the difference of their chemical potentials. Then the following statement is true: if the stability of matter estimate (3) holds, then there exists an interval $]E_a, E_a + \Delta]$ (Δ depends on B) such that for μ in this interval [3]

$$\beta p(\mu, \beta) = \rho_a^{\text{id}}(1 + \mathcal{O}(e^{-c\beta})) \quad \text{as } \beta \rightarrow \infty, \quad c > 0. \quad (8)$$

Namely, if μ is chosen slightly larger than E_a , one obtains the equation of state of a perfect atomic gas up to an exponentially small correction. When $\mu \in]E_a, E_a + \Delta]$, one shows that the following set of inequalities:

$$0 < E_a - 2\mu < E_{N_e N_p} - \mu(N_e + N_p) \quad (9)$$

is satisfied for all $(N_e, N_p) \neq (0, 0), (1, 1)$, where E_{N_e, N_p} is the infimum of the spectrum of H_{N_e, N_p} . In general, the density $\rho_{N_e N_p}^{\text{id}}$ of an ion or molecule made of N_e electrons and N_p protons is defined as

$$\rho_{N_e N_p}^{\text{id}} = d_{N_e N_p} \frac{1}{(2\pi\lambda_{N_e N_p}^2)^{3/2}} e^{-\beta(E_{N_e N_p} - \mu(N_e + N_p))}. \quad (10)$$

Hence, these densities are all exponentially smaller than the atomic density (6).

If μ is lowered below E_a , the energy–entropy balance favors dissociation and the pressure approaches that of an ideal plasma: for $\mu < E_a$

$$\beta p(\mu, \beta) = (\rho_e^{\text{id}} + \rho_p^{\text{id}})(1 + \mathcal{O}(e^{-c\beta})), \quad \beta \rightarrow \infty. \quad (11)$$

3. Correlations and screening

In recent years, a number of exact results have been obtained on the correlations of the Coulomb system at low density. These results are derived in a formal way by the method of quantum Mayer graphs that will be briefly presented in Section 4. They are exact but not rigorous in the sense that they are established for each individual graph, without control of the sum of the diagrammatic series. A review and many references can be found in Ref. [5]. The recombination problem is an old subject which has

also been extensively studied by means of the Green function and Feynman diagram technique of the many-body problem [6].

When we look for more detailed information, we must realize that many different phenomena have a common origin in the basic Coulomb potential. In particular, in the quantum system, there is a variety of possible screening mechanisms. As in the classical Coulomb gas, there is a collective screening effect involving clouds of unbound charges on the scale of the Debye length. At sufficiently low temperature, charges may bind to form neutral atoms or molecules whose diameter is of the order of the Bohr radius. Finally, atomic or molecular dipoles can be polarized to provide dielectric screening.

3.1. The response function to an external classical charge

A localized classical external charge density $c^{\text{ext}}(\mathbf{r})$ gives rise to the additional Coulomb energy

$$U^{\text{ext}} = \sum_{i=1}^N e_{\alpha_i} \int d\mathbf{r} \frac{c^{\text{ext}}(\mathbf{r})}{|\mathbf{r} - \mathbf{r}_i|} \quad (12)$$

to be added to Hamiltonian (1). The linear response function $\tilde{\chi}(\mathbf{k})$ to this perturbation is defined as (in Fourier representation)

$$\tilde{\chi}(\mathbf{k}) = \frac{\tilde{c}^{\text{ind}}(\mathbf{k})}{\tilde{c}^{\text{ext}}(\mathbf{k})} \quad (13)$$

in the limit where $\tilde{c}^{\text{ext}}(\mathbf{k})$, the Fourier transform of $c^{\text{ext}}(\mathbf{r})$, tends to zero. In (13) the induced charge $c^{\text{ind}}(\mathbf{r})$ is the charge density in the presence of the interaction (12). The static dielectric function is related to $\tilde{\chi}(\mathbf{k})$ by

$$\varepsilon(\mathbf{k}) = \frac{1}{\tilde{\chi}(\mathbf{k}) + 1} \quad (14)$$

so $\tilde{\chi}(\mathbf{k})$ yields the basic information about the screening properties. The above formulae hold in a homogeneous fluid phase of the Coulomb system.

3.1.1. Perfect screening

The first statement to be made on $\tilde{\chi}(\mathbf{k})$ is the perfect screening rule in the long wavelength limit, namely,

$$\lim_{k \rightarrow 0} \tilde{\chi}(\mathbf{k}) = -1 \quad (15)$$

for any $\beta < \infty$. This is equivalent to $\lim_{k \rightarrow 0} \varepsilon(\mathbf{k}) = \infty$, showing that a uniform state of the e-p system is a perfect conductor at any positive temperature. This is a consequence of the fact that there is always a tiny fraction of unbound charges, as small as T may be. Relation (15) has been established using the hierarchy equations for the imaginary time Green's functions [7], as well as within the formalism of quantum Mayer graphs [8,9].

3.1.2. Dielectric screening

Then the question arises: under what condition does the e–p system have a dielectric response? Obviously, this will happen in the atomic limit where charges bind to form atomic dipoles. Secondly, in view of (15), the wave number $|\mathbf{k}|$ should not be so small that perfect screening prevails. To determine the range where dielectric behavior occurs, consider the expression of $\tilde{\chi}(\mathbf{k})$ obtained with the mean field Debye–Hückel approximation, neglecting quantum mechanical binding,

$$\tilde{\chi}^{\text{mean field}}(\mathbf{k}) = -\frac{\kappa_{\text{D}}^2}{k^2 + \kappa_{\text{D}}^2}, \quad (16)$$

where

$$\kappa_{\text{D}} = \lambda_{\text{D}}^{-1} = \sqrt{8\pi\beta e^2 \rho_{\text{e}}^{\text{id}}} \quad (17)$$

is the inverse Debye length. On the other hand, the standard dielectric response of a gas of noninteracting hydrogen atoms is

$$\tilde{\chi}^{\text{diel}} = -4\pi\rho_{\text{a}}^{\text{id}}\alpha, \quad (18)$$

where $\alpha = \frac{9}{2}a_{\text{B}}^3$ is the polarizability of a single hydrogen atom submitted to a uniform electric field [10]. Comparing (16) to (18), the dielectric response will dominate if $\tilde{\chi}^{\text{mean field}}(\mathbf{k}) \ll \tilde{\chi}^{\text{diel}}$, implying $k \gg \lambda_{\text{I}}^{-1}$, with

$$\lambda_{\text{I}} = \sqrt{\frac{\alpha}{\beta e^2} \frac{\rho_{\text{a}}^{\text{id}}}{\rho_{\text{e}}^{\text{id}}}} \quad (19)$$

the typical length for the cross-over between ionic and dielectric screening. Moreover, to have the field uniform on the scale of the atomic center of mass distribution, one needs $k \ll \lambda_{\text{a}}^{-1}$ (λ_{a} is the de Broglie thermal length occurring in (6)). Hence, the range of wave numbers k for which (18) is expected to be true is

$$\lambda_{\text{I}}^{-1} \ll k \ll \lambda_{\text{a}}^{-1}. \quad (20)$$

These preliminary considerations being made, one can formulate the result on dielectric screening as follows: choose $\mu \in]E_{\text{a}}, E_{\text{a}} + \Delta]$ in the interval where the atomic phase forms, and let \mathbf{k}_{β} be a sequence of wave numbers such that $\mathbf{k}_{\beta} \rightarrow \mathbf{0}$, $\beta \rightarrow \infty$ with $\mathbf{k}_{\beta}\lambda_{\text{I}} \rightarrow \infty$ and $\mathbf{k}_{\beta}\lambda_{\text{a}} \rightarrow 0$, then [11]

$$\lim_{\beta \rightarrow \infty} \frac{\tilde{\chi}(\mathbf{k}_{\beta})}{\rho_{\text{a}}^{\text{id}}} = -4\pi\alpha. \quad (21)$$

Notice that $\lambda_{\text{I}}^{-1} \sim \exp[\beta(E_{\text{a}} - \mu)/2]$ is exponentially small with β when $\mu > E_{\text{a}}$, but $\lambda_{\text{a}}^{-1} \sim 1/\sqrt{\beta}$. Limit (21) says that for β large, $\tilde{\chi}(\mathbf{k})$ is essentially independent of \mathbf{k} in the range $\lambda_{\text{I}}^{-1} \ll k \ll \lambda_{\text{a}}^{-1}$ and asymptotic to value (18).

3.2. Internal correlations

Some precise asymptotic statements can also be made on the correlations between the particles in the absence of external perturbations. We just quote some results concerning the truncated proton–proton correlation $\rho_{pp}^T(\mathbf{r}_1, \mathbf{r}_2) = \rho_{pp}(\mathbf{r}_1, \mathbf{r}_2) - \rho_p^2$. The first fact is that exponential decay is never true in the quantum gas, but at fixed β and μ (in the fluid phase) [7,8,5]

$$\rho_{pp}^T(\mathbf{r}_1 - \mathbf{r}_2) \sim -\beta \frac{A(\beta, \mu)}{|\mathbf{r}_1 - \mathbf{r}_2|^6} \quad \text{as } |\mathbf{r}_1 - \mathbf{r}_2| \rightarrow \infty. \quad (22)$$

The amplitude A depends on β and μ and can be determined in various asymptotic situations. At fixed β and low density $\mu \rightarrow -\infty$, one finds the explicit formula [12]

$$A(\beta, \mu) \sim -\rho_p^2 \frac{\hbar^4 \beta^3 e^4}{960} \left(\frac{1}{m_e} + \frac{1}{m_p} \right)^2, \quad \mu \rightarrow -\infty \quad (23)$$

showing in particular that, at large distances, the effective potential between two protons in the plasma is attractive! If one considers the atomic limit with $\mu \in]E_a, E_a + \Delta]$, one finds that [13]

$$A(\beta, \mu) \sim (\rho_a^{\text{id}})^2 C_w, \quad \beta \rightarrow \infty, \quad (24)$$

where $C_w < 0$ is the amplitude of the Van der Waals potential between two isolated hydrogen atoms as given in textbooks by a standard second-order perturbation calculation.

4. Quantum Mayer graphs

4.1. The loop Mayer expansion

The mathematical analysis is based on the technique of quantum Mayer graphs [14,5]. These graphs arise when one adopts the Feynman–Kac representation of the Coulomb statistical weight $\exp[-\beta H_{N_e, N_p}]$. Let us recall that for a single particle in a potential V with Hamiltonian $H = \mathbf{p}^2/2m + V$, one has the formula

$$\langle \mathbf{r} | e^{-\beta H} | \mathbf{r} \rangle = \frac{1}{(2\pi\lambda^2)^{3/2}} \int \mathbf{D}(\xi) e^{-\beta \int_0^1 ds V(r+\lambda\xi(s))}, \quad (25)$$

where the integration extends over all the closed Brownian paths $\xi(s)$ (Brownian bridge) such that $\xi(0) = \xi(1) = \mathbf{0}$. Hence a quantum point particle appears as a random charged filament $\lambda\xi(s)$ of extension $\lambda = \hbar\sqrt{\beta/m}$.

With some combinatoric arguments, formula (25) can be extended to the many-body system by regrouping particles of the same charge and same statistics into random charged loops. A loop $\mathcal{L} = (\alpha, q, \mathbf{R}, \mathbf{X}(s))$ is characterized by the particle species α , the number of particles q it contains, its position in space \mathbf{R} and its shape $\mathbf{X}(s)$. $\mathbf{X}(s)$ is again a closed Brownian path parametrized by a “time” s , $0 \leq s \leq q$. The loop carries

a total charge $e_x q$, but also a dipole and higher multipoles generated by the filamentous charge distribution on $\mathbf{X}(s)$. The main advantage of the loop representation is that the statistical mechanics of point quantum charges becomes isomorphic to that of a certain gas of fluctuating multipoles.

The only (but crucial) difference from a genuine classical system is the “equal time” condition. The interaction between two loops inherited from the Feynman–Kac formula is (here for one-particle loops)

$$V(\mathcal{L}_1, \mathcal{L}_2) = \int_0^1 ds V(\mathbf{R}_1 + \lambda_1 \mathbf{X}_1(s) - \mathbf{R}_2 - \lambda_2 \mathbf{X}_2(s)), \quad (26)$$

whereas the electrostatic interaction between two charged wires of shape $\mathbf{X}_1(s)$ and $\mathbf{X}_2(s)$ would be

$$V_{\text{elec}}(\mathcal{L}_1, \mathcal{L}_2) = \int_0^1 ds_1 \int_0^1 ds_2 V(\mathbf{R}_1 + \lambda_1 \mathbf{X}_1(s_1) - \mathbf{R}_2 - \lambda_2 \mathbf{X}_2(s_2)). \quad (27)$$

The lack of Debye screening manifested by the algebraic decay (22) of correlations can precisely be traced out to this difference.

Since averages can be performed according to the rules of classical statistical mechanics in the space of loops, the powerful tools of Mayer diagrams are available. Bonds receive the factor $\exp[-\beta V(\mathcal{L}_i, \mathcal{L}_j)] - 1$ and vertices a renormalized activity $z(\mathcal{L})$ involving the self-energy $U(\mathcal{L})$ of a loop. Coulomb divergencies are still present since $V(\mathcal{L}_1, \mathcal{L}_2) \sim e_1 q_1 e_2 q_2 / |\mathbf{R}_1 - \mathbf{R}_2|$ as $|\mathbf{R}_1 - \mathbf{R}_2| \rightarrow \infty$. They can be cured, as in the classical case, by introducing a screened potential $\phi(\mathcal{L}_a, \mathcal{L}_b)$ by chain diagram resummations. This is the quantum analogue of the Debye potential. We list its principal properties [9].

- (i) $\phi(\mathcal{L}_1, \mathcal{L}_2)$ reduces to the bare Coulomb potential $V(\mathcal{L}_1, \mathcal{L}_2)$ (26) as the density goes to zero.
- (ii) $\phi(\mathcal{L}_1, \mathcal{L}_2)$ (and its powers) is integrable at the origin $\mathbf{R}_1 = \mathbf{R}_2$ because of the smoothing due to the quantum fluctuations.
- (iii) $\phi(\mathcal{L}_1, \mathcal{L}_2) \sim q_1 q_2 \exp[-|\mathbf{R}_1 - \mathbf{R}_2|/\lambda_D] / |\mathbf{R}_1 - \mathbf{R}_2|$ (the classical Debye potential) as $|\mathbf{R}_1 - \mathbf{R}_2| \sim \lambda_D$ (the Debye length).
- (iv) $\phi(\mathcal{L}_1, \mathcal{L}_2)$ is dipolar ($\sim |\mathbf{R}_1 - \mathbf{R}_2|^{-3}$) as $|\mathbf{R}_1 - \mathbf{R}_2| \rightarrow \infty$.

The graphs of the loop Mayer expansion, with resummed ϕ -bonds, are integrable at large distance. They have proven to be very useful to derive low-density expansion of thermodynamics and correlations (see [5] and references therein).

4.2. The screened virial Mayer expansion

The loop formalism is not well suited to the study of the atomic limit because the self-energy $U(\mathcal{L})$ of a loop involves q particles of the same species, therefore not able to bind. By a diagrammatic reorganization, it is possible to convert the loop expansion into the screened virial expansion [15]. In the latter expansion, vertices are labelled by clusters $\mathcal{C}^{(i)} = \mathcal{C}_{N_e N_p}^{(i)}$ of N_e electrons and N_p protons and the associated effective activity

$Z(\mathcal{C}^{(i)})$ comprises all the mutual screened pair interactions of the charges belonging to the cluster with proper quantum statistics. In a gas with short-range forces, the contribution of vertices would yield the standard quantum mechanical virial terms. Here, because of the presence of the screened potential ϕ , vertices are linked by residual bonds $\sim (\Phi(\mathcal{C}^{(i)}, \mathcal{C}^{(j)}))^k$, $k = 1, 2, 3$. The advantage now is that $Z(\mathcal{C})$, after the loop integration, becomes proportional to $\rho_{N_e N_p}^{\text{id}}$ in the atomic limit, hence graphs can be classified according to the ideal densities of (N_e, N_p) clusters. The basic mechanism is as follows. In the atomic limit, ϕ reduces to V so that the loop integrals of $Z(\mathcal{C})$ can be converted to quantum mechanical traces of the type

$$e^{\beta\mu(N_e+N_p)} \text{Tr}\{e^{-\beta H_{N_e N_p}} - \text{“truncation”}\} \quad (28)$$

by the inverse Feynman–Kac formula. In (28), “truncation” means terms that subtract out from $\exp[-\beta H_{N_e N_p}]$ the nonintegrable long-range part, making the trace finite. Moreover, as $\beta \rightarrow \infty$, the dominant contribution to the trace (28) is $\exp[-\beta E_{N_e N_p}]$, with $E_{N_e N_p}$ the ground state energy of $H_{N_e N_p}$, so that (28) is indeed asymptotic to $\rho_{N_e N_p}^{\text{id}}$ (see (10)).

Application of these ideas to the calculation of the response function and the correlations in the atomic limit requires the control of a number of (nonelementary) mathematical points. For instance the dielectric behavior (21) of the response function arises from the graph consisting of a single vertex \mathcal{C}_{11} when k is in the range (20). Then, one has to make sure that one can replace the screened potential by the bare Coulomb potential without creating any divergences. Secondly, one must show that all effects in (28) due to excited and ionized states vanish in this limit. Finally, when μ is in the appropriate range $]E_a, E_a + \Delta]$ where inequalities (9) hold, there must be no contribution of graphs with vertices $\mathcal{C}_{N_e N_p}$, $(N_e, N_p) \neq (1, 1)$. In principle, these techniques can be applied to the classification and evaluation of various Coulomb effects in a general nucleo-electronic plasma at low density.

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