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Chemical potential for the interacting classical gas and the ideal quantum gas obeying a generalized exclusion principle

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Abstract

In this work, we address the concept of the chemical potential μ in classical and quantum gases towards the calculation of the equation of state $\mu = \mu(n, T)$ where n is the particle density and T the absolute temperature using the methods of equilibrium statistical mechanics. Two cases seldom discussed in elementary textbooks are presented with detailed calculations. The first one refers to the explicit calculation of μ for the interacting classical gas exemplified by van der Waals gas. For this purpose, we used the method described by van Kampen (1961 *Physica* **27** 783). The second one refers to the calculation of μ for ideal quantum gases that obey a generalized Pauli's exclusion principle that leads to statistics that go beyond the Bose–Einstein and Fermi–Dirac cases. The audience targeted in this work corresponds mainly to advanced undergraduates and graduate students in the physical–chemical sciences but it is not restricted to them. In regard of this, we have put a special emphasis on showing some additional details of calculations that usually do not appear explicitly in textbooks.

(Some figures may appear in colour only in the online journal)

1. Introduction

Unlike thermodynamic quantities such as temperature T, pressure p or internal energy E, the chemical potential μ has acquired, justified or not, a reputation of a quantity that is not easy to grasp in a physical way. This is particularly true for undergraduate and graduate students

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who take for the first time an introductory course in equilibrium statistical mechanics (ESM). In such courses, micro-canonical, canonical and grand canonical ensembles are introduced in this particular order since the former one only involves counting micro-states compatible with the macroscopic thermodynamic configuration, namely the internal energy. In the second one, the situation changes since it is assumed that the system of interest is in equilibrium with a thermal bath, while in the third one, it is assumed to be in equilibrium with a bath that exchanges energy and particles with the system. Only for the latter system, the chemical potential can be naturally defined because it is a representation where particle exchange is allowed. However, in the thermodynamic limit, all three representations are equivalent. Hence, the question arises as to the true nature of the chemical potential. In this work, we present some ideas that may shed some light on this fundamental issue.

From the micro-canonical and canonical ensembles, the chemical potential can be computed as

$$\mu = -T\left(\frac{\partial S}{\partial N}\right)_{E,V} = \left(\frac{\partial F}{\partial N}\right)_{T,V}, \tag{1}$$

where E, S and $F = E - TS$ denote the internal energy, entropy and the Helmholtz free energy, respectively. In the grand canonical ensemble, μ is assumed to be known as it guarantees the thermodynamic equilibrium of the exchange of particles between the system and the bath. In general, the chemical potential is discussed as a part of a mathematical procedure where minimization of a many-variable distribution function, such as $F(N, V, T)$, plays a fundamental role [1–5]. The chemical potential appears directly in the contribution $\mu\, \mathrm{d}N$ to the change in the internal energy $\mathrm{d}E$ regarding the exchange of particles between the reservoir and the system; thus, μ is the conjugate thermodynamic variable to the number of particles N, given by

$$\mu = \left(\frac{\partial E}{\partial N}\right)_{S,V}, \tag{2}$$

in the same sense as pressure p is a conjugate variable to volume V. Still, a direct physical interpretation in the context of simple gases might be elusive.

A discussion on the meaning of the chemical potential of the ideal gas, based on expression (2), is given in [6], where the authors give physical arguments that attempt to explain the general behaviour of μ as a function of temperature. For the ideal classical gas, μ is a negative quantity given by

$$\mu_{\mathrm{ideal}} = -k_{\mathrm{B}}T\, \ln\left[\frac{V}{N}\left(\frac{mk_{\mathrm{B}}T}{2\pi\hbar^2}\right)^{3/2}\right]. \tag{3}$$

This expression admits an interesting interpretation when the average distance between particles $l \equiv (V/N)^{1/3}$ and the thermal wavelength $\lambda_T = (2\pi\hbar^2/mk_{\mathrm{B}}T)^{1/2}$ are considered. In terms of these, expression (3) can be written as $\mu_{\mathrm{ideal}} = -k_{\mathrm{B}}T\ln\left[l^3/\lambda_T^3\right]$ and the sign of the chemical potential is determined then by the ratio l/λ_T. In the high-temperature limit $\lambda_T \ll l$, where the quantum effects are negligible, μ is a negative quantity (see [6] for an argument on why this is so). In such a situation, the system can be seen as being formed of distinguishable point-particles. In the opposite regime, $\lambda_T \gtrsim l$, the quantum effects are no longer negligible and the wave nature of particles starts to be conspicuous, raising μ to less negative values. This simple reformulation suggests that for the ideal gas, μ can be represented as a balance between length scales which in turn depend on temperature as well[4]. In the low temperature regime, $\lambda_T \gtrsim l$, the dependence of μ on temperature relies on the quantum symmetry under

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[4] It is important to note that even if the possibility of $\mu \geqslant 0$ is open, the model for the classical ideal gas is no longer valid at low temperatures. Thus, for the ideal case, μ is always a negative quantity.

interchange of any pair of particles of the system (indistinguishability). For Fermi particles, μ *decreases monotonically* with temperature from its positive zero-temperature value E_{F} [6] called the Fermi energy which depends on the density of particles as $(N/V)^{2/3}$. The positiveness of E_{F} is a consequence of Pauli's exclusion principle. For Bose particles, μ is also a monotonic decreasing function of temperature (except in the case when a condensate of bosons is possible at a non-zero temperature T_c for which μ is a constant for $T \leqslant T_c$), but decreases from the lowest-lying energy level.

In this paper, we aim to contribute to the understanding of the chemical potential by answering the following question: What are the effects on the chemical potential when interactions among particles are included? For this purpose, we consider the classical van der Waals gas and present the calculation of the chemical potential μ as a function of n and T using the canonical ensemble in ESM. In regard to the quantum case, we consider a generalization of Pauli's exclusion principle and present an explicit calculation, also in the canonical ensemble, of the averaged number of particles $\langle n_k \rangle$ that occupy the single-particle energy level ϵ_k. The calculation presented shows how the chemical potential emerge from the condition that the total number of particles N is conserved. The methodology proposed here can be straightforwardly used in undergraduate and graduate courses on ESM in order to clarify how the concept of chemical potential arises and what is its role in quantum statistics. It is of interest to call the reader's attention to references [7–10], where complementary aspects regarding the chemical potential have been thoroughly discussed.

This work has been organized as follows. In section 2, we present the calculation of the equation of state $\mu = \mu(n, T)$ for the interacting classical gas in terms of the so-called configurational coefficients B_k. Equation $\mu = \mu(n, T)$ contains fundamental information on the thermodynamic properties of a system since it is completely equivalent to the more familiar representation $p = p(V, T)$, where p stands for pressure and V is the volume of the system. A detailed calculation of the correction with respect to the ideal result (3) is made for the van der Waals gas by using the formalism developed by van Kampen [11]. In section 3, we discuss the consequences of a generalization of the Pauli exclusion principle. This generalization explores the possibility that single-particle energy levels ϵ_k of an ideal quantum system may be occupied by at most j particles, where $1 \leqslant j \leqslant \infty$. This generalized statistics is called *intermediate quantum statistics* (IQS). We present our conclusions in section 4 and give some perspectives about the potential applicability of the ideas discussed here. Appendices A and B are included at the end for further references on the treatment of the interacting classical gas and quantum ideal systems displaying IQS.

2. The interacting classical gas

2.1. Generic derivation

It has been shown that the chemical potential for the ideal classical gas is a negative quantity in the temperature regime where quantum effects can be neglected. In order to enhance our intuition on the nature of chemical potential, we will address the calculation of μ for a classical gas where particles interact via a pairwise potential $v(r_1 - r_2)$ using the formalism of ESM. We will use this opportunity to introduce the reader to a method proposed by van Kampen [11] which can be easily extended to consider other types of interactions. To clarify the ideas, we will exemplify the calculation of $\mu(n, T)$ for the van der Waals case. Given the fact that the van der Waals gas is probably one of the most popular cases of study in classical interacting gases, it is of utility to spend some time in getting acquainted with the method.

To take into account the effects of interactions among classical particles on the chemical potential, we will consider the change $\Delta\mu = \mu - \mu_{\mathrm{ideal}}$. This quantity has been related with the averaged work $W(\mathbf{r})$ required to bring an additional particle to the system from infinity to position $\mathbf{r}$ by Widom [14], i.e.

$$\exp\left(-\Delta\mu/k_{\mathrm{B}}T\right) = \langle \exp(-W(\mathbf{r})/k_{\mathrm{B}}T)\rangle, \tag{4}$$

where $\langle ... \rangle$ denotes the average in the canonical ensemble. It seems intuitive to expect $W(\mathbf{r})$ to be larger for a gas with repulsive interactions than for the ideal gas; thus, by using Widom's equivalence (4), we may conjecture that repulsive interactions yield $\Delta\mu > 0$. In general, a system of N interacting particles can be described in ESM by a partition function Z_N of the form [2–4]

$$Z_N = \frac{1}{N!}\left(\frac{mk_{\mathrm{B}}T}{2\pi\hbar^2}\right)^{3N/2} Q_N, \tag{5}$$

where

$$Q_N = \int \mathrm{e}^{-\beta(v_{1,2}+v_{1,3}+\cdots+v_{N-1,N})}\mathrm{d}^3\mathbf{r}_1\cdots\mathrm{d}^3\mathbf{r}_N \tag{6}$$

is known as the configurational integral since it takes into account the different spatial configurations of the system. In equation (6), $v_{i,j} \equiv v(|\mathbf{r}_i - \mathbf{r}_j|)$ is the interaction energy between the ith and jth particles and $\beta = (k_{\mathrm{B}}T)^{-1}$ as usual. A simple method to evaluate Q_N has been given by van Kampen in [11]. In that work, it is suggested that the average of $\mathrm{e}^{-\beta v_{1,2}}\,\mathrm{e}^{-\beta v_{1,3}}\cdots\mathrm{e}^{-\beta v_{N-1,N}}$ over all possible configurations of particle's positions can be identified exactly as the ratio Q_N/V^N. Some mathematical manipulation shows that the configurational partition function Q_N can be expressed as [11]

$$Q_N = V^N \exp\left\{N\sum_{k=1}^{\infty}\left(\frac{N}{V}\right)^k \frac{B_k}{k+1}\right\}, \tag{7}$$

where the coefficients B_k are defined as

$$B_k \equiv \frac{V^k}{k!}\sum_{\{k\}}\int\cdots\int\prod_{i<j}(\mathrm{e}^{-\beta v_{i,j}}-1)\,\mathrm{d}\mathbf{r}_1\cdots\mathrm{d}\mathbf{r}_k, \tag{8}$$

the sum is taken over all *irreducible* terms that involve k-particle position coordinates (see appendix A for more details) and the term $\mathrm{e}^{-\beta v_{i,j}} - 1 = f_{ij}$ is the well-known Mayer function within the context of the Mayer cluster expansion for the partition function of interacting gases [12]. Such an expansion of the partition function is a representation of the interactions of a real gas in terms of sets of clusters of particles of a given size. The higher the term in the expansion, the higher the number of clusters considered [13]. The total partition function Z_N is then given in terms of the coefficients B_k as

$$Z_N = \frac{V^N}{N!}\left(\frac{mk_{\mathrm{B}}T}{2\pi\hbar^2}\right)^{3N/2}\exp\left\{N\sum_{k=1}^{\infty}\left(\frac{N}{V}\right)^k\frac{B_k}{k+1}\right\}, \tag{9}$$

and the Helmholtz free energy F by

$$F = -Nk_{\mathrm{B}}T\ln\left[\frac{V}{N}\left(\frac{mk_{\mathrm{B}}T}{2\pi\hbar^2}\right)^{3/2}\right] - Nk_{\mathrm{B}}T\left[1+\sum_{k=1}^{\infty}\left(\frac{N}{V}\right)^k\frac{B_k}{k+1}\right]. \tag{10}$$

The chemical potential μ can thus be obtained as

$$\mu = \mu_{\mathrm{ideal}} - k_{\mathrm{B}}T\sum_{k=1}^{\infty}\left(\frac{N}{V}\right)^k B_k, \tag{11}$$

where μ_{ideal} stands for the expression obtained in equation (3) and B_k are the coefficients in equation (8). Once the specific functional form for the interaction potential $v(r_1 - r_2)$ is given, all B_ks can be calculated in principle.

2.2. Specific example: the van der Waals gas

Expression (11) is a general result. It gives μ for the classical interacting gas as a series of powers in the particle density $(N/V)^k$ and the coefficients B_k. The immediate consequence here is that interactions shift the value of the chemical potential from the ideal case. In spite of its generality, in practice, the calculation of B_k for $k > 2$ is only analytically possible for a small set of interaction potentials. However, for enough dilute systems, i.e. $N/V \ll 1$, we may consider only the first term of equation (11) as a valid approximation. Thus, at first order in N/V, we have $\mu = \mu_{\text{ideal}} - k_B T(N/V)B_1$, where B_1 depends on the specific interatomic potential between particles. In order to obtain quantitative results about the effects of interactions on the chemical potential, let us consider the van der Waals gas as a specific example.

One of the most commonly used pairwise potentials that approximates the well-known semi-empirical Lennard–Jones potential $v(r) = v_0[(d/r)^{12} - 2(d/r)^6]$ can be written as

$$v(r) = \begin{cases} \infty & \text{for } r < d \\ -v_0(d/r)^6 & \text{for } r \geqslant d, \end{cases} \tag{12}$$

where v_0 is the minimum interaction energy between a pair of particles and d their separation for that value of energy. For this model, B_1 can be evaluated exactly as follows. Spherical symmetry enables us to write $B_1 = \int (\mathrm{e}^{-\beta v(\mathbf{r})} - 1)\,\mathrm{d}\mathbf{r} = 4\pi \int_0^\infty r^2(\mathrm{e}^{-\beta v(r)} - 1)\,\mathrm{d}r$. Then, by splitting the last integral into one integral from 0 to d plus a second one from d to ∞ and using the fact that $v(r) \to \infty$ for $0 < r < d$, we get

$$B_1 = 4\pi \left[\int_d^\infty (\mathrm{e}^{\beta v_0 (d/r)^6} - 1) r^2\,\mathrm{d}r - \frac{d^3}{3} \right]. \tag{13}$$

The integral in equation (13) can be evaluated directly by using the Taylor series of the exponential function. After integrating term by term, we obtain

$$B_1 = \frac{4}{3}\pi d^3 \left[\sum_{n=1}^{\infty} \frac{(\beta v_0)^n}{(2n-1)n!} - 1 \right]. \tag{14}$$

It is possible to go a step further in order to write equation (14) in terms of elementary functions. The infinite sum in equation (14) can be expressed as $\sum_{n=1}^{\infty} x^n/(2n-1)n! = 1 - \mathrm{e}^x + (\pi x)^{1/2}\mathrm{erfi}(x^{1/2})$, where $\mathrm{erfi}(z) = -\mathrm{i}\,\mathrm{erf}(\mathrm{i}z)$ denotes the imaginary error function. Thus, a simple expression for the correction factor $\Delta\mu \equiv \mu - \mu_{\text{ideal}}$ can be obtained for temperatures such that $k_B T \gg v_0$, since only the first term in the series expansion in expression (14) is needed. With these considerations and recalling that $l = (V/N)^{1/3}$, we have

$$\Delta\mu \simeq k_B T \frac{4}{3}\pi \left(\frac{d}{l}\right)^3 \left(1 - \frac{v_0}{k_B T}\right) > 0 \tag{15}$$

in agreement with Monte Carlo calculations obtained previously by other authors [15].

For relatively high temperatures, $v_0/k_B T \approx 0$ and then it is the hardcore repulsion of the inter-particle interaction which governs the dynamics of the gas. In this limit, the system corresponds to a hard-sphere gas, thus giving $\Delta\mu = k_B T \frac{4}{3}\pi(d/l)^3$ in this case [15]. For temperatures smaller than v_0/k_B, $\Delta\mu$ becomes negative (see figure 1), but this should not be considered correct since at such temperatures we are out of the classical regime and quantum corrections must be taken into account. In terms of the parameters a and b of the standard van der Waals equation of state

$$\left(p + \frac{N^2}{V^2}a\right)(V - Nb) = Nk_B T, \tag{16}$$

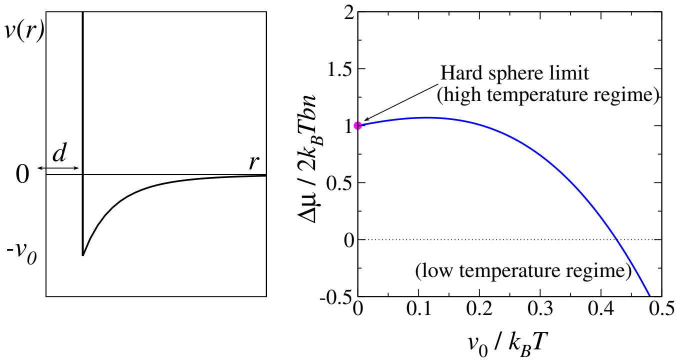

Figure 1. Left: the model potential given by expression (12) as an approximation to the more realistic Lennard–Jones potential. Right: the change in the chemical potential, given by (15), as a function of the ratio of the energy that characterizes the interacting potential v_0 to the thermal energy.

Table 1. Values of the van der Waals parameters a and b for some substances are given. With these values, the ratio $v_0/k_{\mathrm{B}}T_R$ is computed, where T_R denotes the room temperature.

Substance	a	b	$v_0/k_{\mathrm{B}}T_R$
Helium	0.0346	0.0238	0.0603
Neon	0.208	0.0167	0.516
Hydrogen	0.2452	0.0265	0.384
Oxygen	1.382	0.0319	1.796
Water	5.537	0.0305	7.527

the chemical potential for the van der Waals gas can be written as

$$\mu_{vdw} = \mu_{\mathrm{ideal}} - 2\left(\frac{N}{V}\right)(a - k_{\mathrm{B}}Tb)\,, \tag{17}$$

where $a = v_0 b = v_0 \frac{2}{3}\pi d^3$. Table 1 presents some standard values for a and b parameters for different gases [16]. We encourage the interested reader to get immersed into the details of the calculations by using another interacting potentials v or trying to calculate analytically or numerically higher order corrections to the result obtained here.

General thermodynamic principles for single-component systems ensure that only two independent variables are needed to describe completely the state of the system. Thus, $\mu = \mu(n, T)$ and $p = p(V, T)$ are both valid and equivalent representations. The relevance of calculating $\mu = \mu(n, T)$ is then twofold. On one hand, this type of elementary calculation in simple systems enables us to gain some confidence in the methods of ESM that may guide us to more complex situations. On the other hand and in a more physical arena, the knowledge of $\mu = \mu(n, T)$ opens up the door to the analysis and the description of the phase transitions inherent to the system under study.

3. Generalized quantum statistics: beyond Bose–Einstein and Fermi–Dirac

At low temperatures, gases depart from classical behaviour since quantum wave-like properties of matter emerge. One of the main classical properties washed away in the quantum regime is distinguishability. In the classical picture, we can in principle tag and follow the trajectory of any particle; however, this is not the case in the quantum regime [17]. This property has profound consequences in the number Ω of different micro-states available to the system and consequently in the macroscopic properties of the system according to the fundamental relation $S = k_B \log \Omega$ proposed by Boltzmann. In general, classical systems will tend to exhibit more micro-states since permutations among particles result in different configurations due to distinguishability. Quantum systems, on the other hand, display a smaller number of different configurations. As discussed before, the thermal wavelength λ_T serves as a length scale over which wave-like behaviour appears. For high temperatures, $\lambda_T \rightarrow 0$ and then the particles can be visualized as classical point-like particles with a definite momentum and position. However, as temperature is lowered, λ_T starts to increase accordingly down to a characteristic temperature T^*, such that the wavelength of particles is of the same order of magnitude as the average distance l between any two particles. At this temperature T^*, the system enters into the so-called *degeneracy regime*.

Two quantum effects emerge due to indistinguishability. The first case corresponds to systems formed by particles called bosons whose spin is an integer multiple of $\hbar$. The second one refers to systems formed by fermions whose spin is a half-integer multiple of $\hbar$. This relation between spin and statistics was established by Pauli [18]. These systems, Bose and Fermi gases, exhibit completely different macroscopic properties, in particular regarding the chemical potential. As is presented in standard textbooks [2–4], Bose–Einstein (BE) and Fermi–Dirac (FD) statistics correspond to two opposite regimes ruled by Pauli's exclusion principle. This fundamental principle constrains the occupancy of a particular energy level to the value 0 or 1 in the case of FD statistics and no restriction at all in the BE case, leaving aside the discussion on the possibility of considering an intermediate case between BE and FD statistics. In order to gain more intuition with respect to the chemical potential in an ideal quantum system, we discuss in this section a generalization of Pauli's exclusion principle related to what we have called IQS.

In principle, it is possible to define a system to have what we have called IQS. In such a case, the averaged occupancy $\langle n_k \rangle$, i.e. the average number of particles allowed to occupy a particular energy level k, is not restricted to the values for the Bose or the Fermi gases. Let us consider a quantum ideal gas composed of N indistinguishable spinless particles that can be distributed along a set of $\{\epsilon_k\}$ energy levels. Let us consider the case in which the number of particles is conserved. Also consider the situation in which due to indistinguishability, each energy level ϵ_k can be populated, at most, by j particles, with $j = 1, 2, \ldots, \infty$. The variable j denotes the order of the IQS and we will refer to this as IQS$_j$. This generalizes Pauli's exclusion principle in such a way that it interpolates between the BE case with $j = \infty$ and the FD case corresponding to $j = 1$. We now calculate the average number of particles $\langle n_k \rangle_j$ that occupy the single-particle energy k in a system subject to the IQS$_j$. The calculation presented here is carried out by using the canonical ensemble; thus, we focus on the calculation of the partition function of N particles $Z_N = \sum_{\{n_k\}} \mathrm{e}^{-\beta E_{\{n_k\}}}$, where $\{n_k\}$ denote the possible configurations for a particular distribution of n_1 particles in the energy level ϵ_1, n_2 particles in the level ϵ_2 and so on, compatible with the restrictions

$$\sum_k n_k = N, \tag{18}$$

$n_k = 0, 1, \ldots, j$ given by the statistics, and $E_{\{n_k\}} = \sum_k n_k \epsilon_k$ being the total energy associated with the particular configuration of n_ks.

It is possible to calculate the average number of particles $\langle n_k \rangle_j$ occupying the single-particle energy k for an ideal quantum gas exhibiting IQS of order j by using the method proposed in appendix B. We strongly suggest that the reader follows the technical details presented there. The final result allows us to write the average occupancy of the energy level k for a quantum system with an IQS of order j in a closed formula as

$$\langle n_k \rangle_j = \frac{1}{\mathrm{e}^{\beta(\epsilon_k-\mu)}-1} - \frac{j+1}{\mathrm{e}^{\beta(\epsilon_k-\mu)(j+1)}-1}. \tag{19}$$

Equation (19) provides evidence that the chemical potential μ is always associated with particle conservation requirements in ideal quantum gases displaying IQS of any order.

Special cases. For $j = 1$, equation (19) reduces to the well-known result for the Fermi–Dirac statistics:

$$\langle n_k \rangle_{\mathrm{FD}} = \frac{1}{\mathrm{e}^{\beta(\epsilon_k-\mu)}+1}. \tag{20}$$

The limit $j \to \infty$ is well defined always so that $\mu \leqslant \epsilon_k$ for all values of k. This limit leads to the BE distribution

$$\langle n_k \rangle_{\mathrm{BE}} = \frac{1}{\mathrm{e}^{\beta(\epsilon_k-\mu)}-1}. \tag{21}$$

From equation (19), it is possible to obtain closed and analytical expressions for the average occupancy of a system obeying an IQS of any order. For instance, for $j = 2$, each energy level ϵ_k may be occupied by zero, one or two particles, and $\langle n_k \rangle_2$ is then given by

$$\langle n_k \rangle_2 = \frac{\mathrm{e}^{-\beta(\epsilon_k-\mu)} + 2\mathrm{e}^{-2\beta(\epsilon_k-\mu)}}{1 + \mathrm{e}^{-\beta(\epsilon_k-\mu)} + \mathrm{e}^{-2\beta(\epsilon_k-\mu)}}. \tag{22}$$

Any other value for $\langle n_k \rangle_j$ can be obtained properly from the general result given above.

3.1. The equation of state $\mu = \mu(n, T)$

The behaviour of the equation of state $\mu = \mu(n, T)$, with $n = N/V$, is given by the implicit relation

$$N = \sum_k \langle n_k \rangle_j = \sum_k \left[\frac{1}{\mathrm{e}^{\beta(\epsilon_k-\mu)}-1} - \frac{j+1}{\mathrm{e}^{\beta(\epsilon_k-\mu)(j+1)}-1} \right]. \tag{23}$$

If the spacing between energy levels is small compared to $k_{\mathrm{B}}T$ or in the thermodynamic limit, we can replace the sum by an integral, i.e.

$$n = \int_0^\infty d\epsilon\, \rho(\epsilon) \left[\frac{1}{\mathrm{e}^{\beta(\epsilon-\mu)}-1} - \frac{j+1}{\mathrm{e}^{\beta(\epsilon-\mu)(j+1)}-1} \right], \tag{24}$$

where $\rho(\epsilon)$ is the density of states which depends on both the system itself and its dimensionality [17]. For the free particle in three dimensions, we have

$$\rho(\epsilon) = \frac{2}{\pi^{1/2}} \left(\frac{m}{2\pi\hbar^2} \right)^{3/2} \epsilon^{1/2} \tag{25}$$

with m the mass of the particles. Expression (24) can then be written as

$$n = \left(\frac{mk_{\mathrm{B}}T}{2\pi\hbar^2} \right)^{3/2} \left[Li_{3/2}(\mathrm{e}^{\beta\mu}) - \frac{1}{(j+1)^{1/2}} Li_{3/2}(\mathrm{e}^{\beta\mu(j+1)}) \right], \tag{26}$$

where $Li_s(\zeta)$ is the poly-logarithm function defined as

$$Li_s(\zeta) = \frac{1}{\Gamma(s)} \int_0^\infty \frac{t^{s-1}\,\mathrm{d}t}{\zeta^{-1}\,\mathrm{e}^t - 1}. \tag{27}$$

It is not obvious or straightforward to see whether the chemical potential is a positive or negative quantity only from expression (24); however, in the limit of high temperature, we expect $\langle n_k \rangle_j \ll 1$ and from equation (19) we have the classical result that $\mu < 0$. By solving numerically equation (24), the monotonic-decreasing dependence on the temperature of μ is apparent in the left panel of figure 2 for $j = 1, 2, 8, 16, 32$ and 128. For the same values, the isothermal dependence on the density of μ is shown in the right panel.

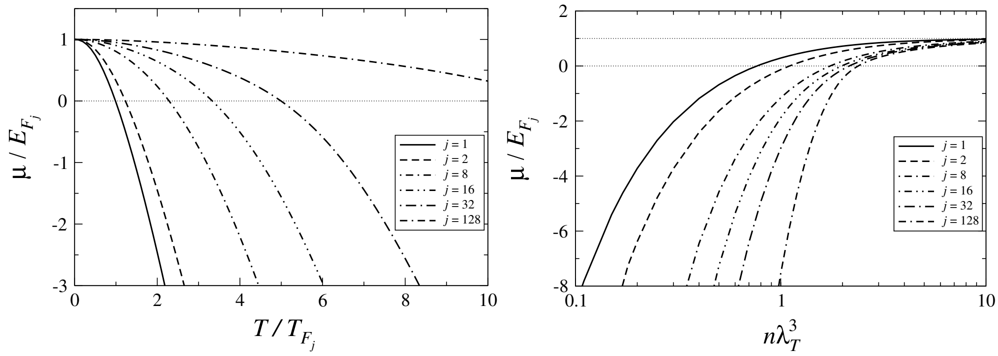

Figure 2. Chemical potential, normalized with $E_{\mathrm{F}j}$ (28), is presented as a function of the dimensionless temperature $T/T_{\mathrm{F}j}$ in the left panel for different values of the order j of the statistics. $T_{\mathrm{F}j}$ is defined by $E_{\mathrm{F}j}/k_{\mathrm{B}}$. Right: the dependence on the dimensionless density $n\lambda_T^3$ of $\mu/E_{\mathrm{F}j}$ is presented. In both cases, we have chosen $j = 1$, 2, 8, 16, 32 and 128, the former corresponding to the chemical potential of the ideal Fermi gas.

The chemical potential at $T = 0$. At zero temperature, one expects that the first energy levels, up to the generalized Fermi energy, $E_{\mathrm{F}j}$, are being occupied by exactly j particles. For this to hold, it is necessary that in expression (B.13), $\epsilon - \mu < 0$ otherwise $\langle n_\epsilon \rangle = 0$, leading to $\mu = E_{\mathrm{F}j}$. By putting $T = 0$ in (24), we have for the three-dimensional free gas that

$$E_{\mathrm{F}j} = \frac{\hbar^2}{2m}\left[\frac{6\pi^2}{j}n\right]^{2/3}, \tag{28}$$

which reduces to the usual Fermi energy E_{F} of a spinless ideal Fermi gas when $j = 1$ (the dependence on n is shown in figure 3 by the dotted line) and it vanishes in the limit $j \to \infty$ of the BE statistics. It is clear that the change in energy by adding only one particle to the system corresponds to the chemical potential (there is no change in entropy at zero temperature). For $j = 1$, $\mu = E_{\mathrm{F}} > 0$ and starts to diminish as j increases, vanishing in the limit $j \to \infty$, reflecting the nature of the statistical potential interaction introduced in [2] for the BE and FD cases.

Special cases. For the BE and FD statistics, the equation of state (26) can be described in a unified way by using the polylogarithm function [20] in the following way:

$$n = \pm\left(\frac{mk_{\mathrm{B}}T}{2\pi\hbar^2}\right)^{3/2} Li_{3/2}(\pm \mathrm{e}^{\beta\mu}), \tag{29}$$

which corresponds to the equation of state $\mu = \mu(n, T)$ for the ideal Bose (+) and Fermi (−) gas, respectively. This is completely equivalent to the standard equation of state for particle density n in terms of volume V and pressure p; see, for example, [21]. Both $\mu = \mu(n, T)$ and $n = n(T, p)$ contain the same information and thus can be used indistinctly to obtain the thermodynamic information of the system. In figure 3, the monotonic dependence on temperature of the chemical potential is shown for a fixed value of particle density in the left panel and as a function of the particle density for various isotherms in the right one. An analytical expression for $\mu(T)$ can be given for the Fermi gas in the regime $T/T_{\mathrm{F}} \ll 1$, where

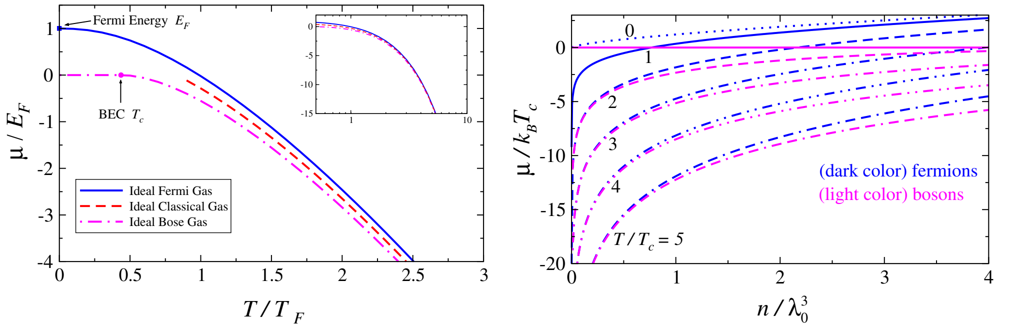

Figure 3. Left: chemical potential in units of E_{F} (the Fermi energy of a spinless ideal Fermi gas) as a function of temperature in units of $T_{\mathrm{F}} = E_{\mathrm{F}}/k_{\mathrm{B}}$, k_{B} being Boltzmann's constant, for (i) the ideal Fermi gas (continuous-blue/dark grey line), (ii) the ideal classical gas (dashed line) and (iii) the ideal Bose gas (dash-dotted line). The BEC critical temperature $T_c/T_{\mathrm{F}} = [4/(3\zeta(3/2)2\sqrt{2})]^{2/3} \simeq 0.436$ is marked with a dot and the Fermi energy with a blue square. The inset shows how the chemical potential of the ideal quantum gas approaches the classical one at large temperatures. Right: chemical potential μ in units of $k_{\mathrm{B}}T_c$ for the ideal Bose (light lines) and Fermi gases (dark lines) as a function of the particle density n for different values of the dimensionless temperature $T/T_c = 0.0,\ 1.0,\ 2.0,\ 3.0,\ 4.0,\ 5.0$. T_c corresponds to the BE condensation critical temperature (equation (31)) of a boson gas with arbitrary density n_0. The variable λ_0 corresponds to the thermal wavelength evaluated at $T = T_c$. Note that both cases converge to the same values of the chemical potential for small enough density. This corresponds to the classical limit.

the Fermi temperature T_{F} is defined by $E_{\mathrm{F}}/k_{\mathrm{B}}$. In this regime, the well-known Sommerfeld approximation (see [22] for details) gives

$$\mu_{\mathrm{F}} = E_{\mathrm{F}}\left[1 - \frac{\pi^2}{12}(T/T_{\mathrm{F}})^2 + \cdots\right], \tag{30}$$

which shows that the chemical potential for the Fermi gas, μ_{F}, decreases from its maximum value E_{F}. The temperature $\tilde{T}$ that separates the $\mu > 0$ region from the $\mu < 0$ one can be computed exactly and is given by $\tilde{T} = [\Gamma(5/2)\zeta(3/2)(1 - \sqrt{2}/2)]^{-2/3}\, T_{\mathrm{F}} \simeq 0.989\, T_{\mathrm{F}}$. Note that the transition to the classical behaviour can occur at very high temperatures, as high as T_{F} which for a typical metal is of the order of 10^4 K. The isotherms shown in the right panel of figure 3 show the classical behaviour at low densities.

For the ideal Bose gas, a different picture emerges. First, since $\langle n_k \rangle$ is a non-negative quantity, it is required that $\epsilon_j - \mu \leqslant 0$ for all j, which implies that $\mu \leqslant \epsilon_0$, where ϵ_0 is the single-particle ground-state energy. In general, $\epsilon_0 \to 0$ in the thermodynamic limit, leading to the well-known result $\mu \leqslant 0$ for all temperatures. Furthermore, as discussed in many textbooks [2, 3], a phase transition occurs at a critical temperature $T_c > 0$ for the free three-dimensional boson gas. Such transition temperature is given by

$$T_c = \frac{2\pi}{\zeta(3/2)}\frac{\hbar^2}{k_{\mathrm{B}}m}n^{2/3} \tag{31}$$

marked with a dot in the left panel of figure 3, and its value in units of T_{F} is given by $T_c/T_{\mathrm{F}} = [4/(3\zeta(3/2)2\sqrt{2})]^{2/3} \simeq 0.436$. Expression (31) is obtained from equation (29) when the conditions $\mu(T_c) = 0$ and $\langle n_{\epsilon=0}\rangle = 0$ are set. In expression (31), the quantity $\zeta(3/2) = \mathrm{Li}_{3/2}(1)$ is the zeta function of Riemann. In figure 3, isothermal curves (light colour) of $\mu(n, T)$ are shown. The critical density n_c at which BEC occurs is determined by $\mu(n_c, T) = 0$ and is given by $n_c = (mk_{\mathrm{B}}T/2\pi\hbar^2)^{2/2}\zeta(3/2)$ where all the symbols have been defined previously.

4. Conclusions and perspectives

We have addressed the behaviour of the chemical potential μ for the interacting classical gas and the ideal quantum gas that obeys a generalized exclusion principle. For the former case, we used a method proposed by van Kampen [11] to evaluate the partition function of the interacting classical gas. The calculation allowed us to find the equation of state $\mu = \mu(n, T)$ for a pairwise interaction potential in the case of the van der Waals gas. We observed a positive shift $\Delta\mu$ with respect to the ideal gas due, basically, to the repulsive nature of the interactions as was conjectured using Widom's equivalence. In contrast, as shown in figure 1, if temperature is kept constant and v_0 is increased making the interaction more attractive, the chemical potential lowers its value with respect to the ideal case leading to a negative shift.

Another novel feature discussed here refers to the introduction of quantum statistics beyond the standard BE and FD cases. These statistics that we have called IQS enable us to treat quantum ideal systems with single-particle energy levels occupancies $\langle n_k \rangle$ between zero up to j particles including the $j = \infty$ case. We provided an explicit calculation using the canonical ensemble to obtain the most general form for the averaged number of particles $\langle n_k \rangle_j$, that occupy the energy level ϵ_k, and its relation with the chemical potential. The procedure shown here and in particular the one that refers to equation (B.11) can be of some use to discern between the results given by the canonical and grand canonical ensembles studied for the fermionic lattice gas in [23]. We also showed that this general result contains the particular cases of BE and FD statistics. Quantum systems exhibiting *fractional statistics* are not just an academic exercise; quasi-particles called *anyons* display this property [24, 25]. In two-dimensional systems, these quasi-particles have found application in the theory of the quantum Hall effect and anyon superconductivity [26]. Emphasis in the equation of state, $\mu = \mu(n, T)$, has been placed, since it encodes substantial information regarding the existence of phase transitions in the system and is completely equivalent to the more familiar representation $p = p(V, T)$, with p and V the system pressure and volume, respectively.

Finally, as open problems that could be carried out in a standard course on ESM, we propose the calculation of the shift of the chemical potential for the non-ideal classical gas if the interactions are short ranged and attractive, the calculation of the statistical potential (as is done in [2] for the BE and FD statistics) for the IQS$_j$ and a discussion on the case when N is not conserved, as happens in many quasi-particle systems.

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Appendix A. Evaluation of the configurational integral Q_N

The basic idea to evaluate the configurational integral Q_N given by equation (6) is to compute the statistical average of $\overline{\mathrm{e}^{-\beta v_{1,2}}\, \mathrm{e}^{-\beta v_{1,3}} \cdots \mathrm{e}^{-\beta v_{N-1,N}}}$ over all possible configurations of the particle positions denoted with $Q_N/V^N = \overline{\mathrm{e}^{-\beta v_{1,2}}\, \mathrm{e}^{-\beta v_{1,3}} \cdots \mathrm{e}^{-\beta v_{N-1,N}}}$. van Kampen's approach is based on a factorization of D into terms of D_k that takes into account the correlations of $k \geqslant 2$ particles, i.e.

$$Q_N/V^N = \prod_{k=2}^{N}(d_k), \tag{A.1}$$

which gives the number of combinations of k particles taken from the total N, and

$$d_k = \frac{\overline{\mathrm{e}^{-\beta v_{1,2}}\mathrm{e}^{-\beta v_{1,3}}\ldots\mathrm{e}^{-\beta v_{k-1,k}}}}{D}, \tag{A.2}$$

with D the immediate lower approximation for the same numerator.

For $k = 2$, $d_2 = \overline{\mathrm{e}^{-\beta v_{1,2}}}$ since $D = 1$ in this case. Thus, the first factor in equation (A.1) is given by

$$\overline{\mathrm{e}^{-\beta v_{1,2}}}^{N(N-1)/2} = \left[V^{-1}\int \mathrm{d}\mathbf{r}_1 V^{-1}\int \mathrm{d}\mathbf{r}_2\,\mathrm{e}^{-\beta v_{1,2}}\right]^{N(N-1)/2}. \tag{A.3}$$

In order to take the thermodynamic limit $N, V \to \infty$ with N/V constant, consider the following identity:

$$\left[\int\frac{\mathrm{d}\mathbf{r}_1}{V}\int\frac{\mathrm{d}\mathbf{r}_2}{V}\,\mathrm{e}^{-\beta v_{1,2}}\right]^{(N-1)/2} = \left[1+\frac{1}{N}\frac{N}{V}\int \mathrm{d}\mathbf{r}(\mathrm{e}^{-\beta v(\mathbf{r})}-1)\right]^{(N-1)/2}, \tag{A.4}$$

thus giving as a result $d_2^{\binom{N}{2}} = \exp\left\{\frac{N^2}{2V}B_1\right\}$ with $B_1 \equiv \int \mathrm{d}\mathbf{r}\,(\mathrm{e}^{-\beta v(\mathbf{r})}-1)$. For dilute enough systems where only correlations of two particles are important, this approximation should work fine. The calculation of the general factor $(d_k)^{\binom{N}{k}}$ is more involved and we only present a sketch of it. By writing $\mathrm{e}^{-\beta v_{i,j}} = 1 + f_{i,j}$, equation (A.2) can be rewritten as

$$d_k = \frac{1+\overline{f_{1,2}}+\cdots+\overline{f_{1,2}f_{1,3}\cdots f_{k-1,k}}}{D}. \tag{A.5}$$

van Kampen argues that the class of terms in the numerator of (A.5) that involve less than k particles and those that involve k particles but are reducible are also present in D, such that the numerator can be written as $\big(1+\sum_{\{k\}}\overline{f_{1,2}f_{1,3}\cdots}+\mathcal{O}(V^{-k})\big)D$, where the summation extends over all irreducible terms that involve $2,\ldots,k$ particles. A term of the form $\int\cdots\int\prod_{i<j}g_{i,j}\,\mathrm{d}\mathbf{r}_1\cdots\mathrm{d}\mathbf{r}_k$, with $g_{i,j}$ an arbitrary function of $|\mathbf{r}_i-\mathbf{r}_j|$, is said to be irreducible if it cannot be factorized into products of integrals of $g_{i,j}$ involving less than k particle-position coordinates. For instance, it is straightforward to check that $\iiint \mathrm{d}\mathbf{r}_1\,\mathrm{d}\mathbf{r}_2\,\mathrm{d}\mathbf{r}_3\,g_{1,2}g_{1,3}g_{2,3}$ is irreducible while the integral $\iiint \mathrm{d}\mathbf{r}_1\,\mathrm{d}\mathbf{r}_2\,\mathrm{d}\mathbf{r}_3\,g_{1,2}g_{1,3} = V\big[\int \mathrm{d}\mathbf{r}g(\mathbf{r})\big]^2$ is not; here we have used the identity $\int \mathrm{d}\mathbf{r} = V$.

Thus, we have

$$d_k = 1+\frac{(k-1)!}{V^{k-1}}B_{k-1}+\mathcal{O}(V^{-k}), \tag{A.6}$$

where B_k is given by (8) and we have recognized $\sum_{\{k\}}\overline{f_{1,2}f_{1,3}\ldots}$ with the usual irreducible cluster integral [2–4] $\frac{(k-1)!}{V^{k-1}}B_{k-1}$. In the thermodynamic limit, the factor $(d_k)^{\binom{N}{k}}$ can then be written as

$$\exp\left\{\frac{N^k}{V^{k-1}}\frac{B_{k-1}}{k}\right\}$$

and by combining this result with the result for $k = 2$, we finally get the desired result given by expression (7).

Appendix B. Derivation of mean occupancy for an ideal quantum gas with intermediate quantum statistics of order *j*

Our starting point to derive $\langle n_k\rangle_j$ is the observation that Z_N can be written as a sum over all possible values of n_k for each k, i.e.

$$Z_N = \sum_{n_1=0}^{j}\sum_{n_2=0}^{j}\cdots \mathrm{e}^{-\beta(n_1\epsilon_1+n_2\epsilon_2+\cdots)}\delta_{n_1+n_2+\cdots,N}, \tag{B.1}$$

where $\delta_{i,j}$ is the Kronecker delta and the factor $\delta_{n_1+\cdots+n_k+\cdots,N}$ has been introduced to guarantee the conservation of N. To proceed further [4], we write the last expression as

$$Z_N = \sum_{n_k=0}^{j}\mathrm{e}^{-\beta n_k\epsilon_k}\left[\sum_{n_1=0}^{j}\sum_{n_2=0}^{j}\cdots \mathrm{e}^{-\beta(n_1\epsilon_1+n_2\epsilon_2+\cdots)}\delta_{n_1+n_2+\cdots,N-n_k}\right] \tag{B.2}$$

$$= \sum_{n_k=0}^{j}\mathrm{e}^{-\beta n_k\epsilon_k}\left[\sum_{\{n_l\}}{}'\mathrm{e}^{-\beta E'_{\{n_l\}}}\delta_{n_1+n_2+\cdots,N-n_k}\right]. \tag{B.3}$$

The term within square brackets corresponds to the partition function of $N-n_k$ particles with the energy level k excluded, $Z^{(k)}_{N-n_k}$; we use the superindex ${}^{(k)}$ to denote all quantities which have been computed in this way. In terms of this, we have

$$Z_N = \sum_{n_k=0}^{j}\mathrm{e}^{-\beta n_k\epsilon_k}Z^{(k)}_{N-n_k}. \tag{B.4}$$

A general formula to compute average number of particles that occupy the energy level k, given by $\langle n_k\rangle_j = Z_N^{-1}\sum_{n_1=0}^{j}\sum_{n_2=0}^{j}\cdots n_k\mathrm{e}^{-\beta(n_1\epsilon_1+n_2\epsilon_2+\cdots)}$, is derived by noting that

$$\langle n_k\rangle_j = -\frac{1}{\beta}\frac{\partial \ln Z_N}{\partial\epsilon_k}; \tag{B.5}$$

thus,

$$\langle n_k\rangle_j = \frac{1}{Z_N}\sum_{n_k=0}^{j} n_k\mathrm{e}^{-\beta n_k\epsilon_k}Z^{(k)}_{N-n_k}. \tag{B.6}$$

The evaluation of equation (B.6) requires us to compute $Z^{(k)}_{N-n_k}$ from $n_k = 0$ to j which makes the calculation rather cumbersome (see [19] and [4] for details). We avoid this difficulty by noting that the ratio $Z^{(k)}_{N-n_k}/Z^{(k)}_N$ can be written as the product of the ratios of partition functions that differ only in one particle, i.e.

$$\frac{Z^{(k)}_{N-n_k}}{Z^{(k)}_N} = \frac{Z^{(k)}_{N-1}}{Z^{(k)}_N}\cdot\frac{Z^{(k)}_{N-2}}{Z^{(k)}_{N-1}}\cdots\frac{Z^{(k)}_{N-n_k}}{Z^{(k)}_{N-n_k+1}}. \tag{B.7}$$

Consider the energy of $N-n_k$ particles distributed over the energy levels distinct to ϵ_k

$$E^{(k)}_{N-n_k} = \frac{\sum_{\{n_l\}}{}'E'_{\{n_l\}}\mathrm{e}^{-\beta E'_{\{n_l\}}}\delta_{n_1+n_2+\cdots,N-n_k}}{\sum_{\{n_l\}}{}'\mathrm{e}^{-\beta E'_{\{n_l\}}}\delta_{n_1+n_2+\cdots,N-n_k}} = -\frac{\partial}{\partial\beta}\ln Z^{(k)}_{N-n_k}. \tag{B.8}$$

The difference $\Delta E^{(k)} = E^{(k)}_{N-n_k} - E^{(k)}_{N-n_k+1}$, i.e. the change in energy when withdrawing only one particle, is given by

$$\Delta E^{(k)} = -\frac{\partial}{\partial\beta}\ln\frac{Z^{(k)}_{N-n_k}}{Z^{(k)}_{N-n_k+1}}. \tag{B.9}$$

Since the energy change has been done at constant T and V, we must have that $\beta\Delta E^{(k)} + \ln\left[Z^{(k)}_{N-n_k}/Z^{(k)}_{N-n_k+1}\right] = \Delta S^{(k)}/k_{\rm B}$, where $\Delta S^{(k)}$ denotes the entropy change of the system when withdrawing only one particle.

Using the first law of thermodynamics, we can identify the chemical potential with

$$\mu^{(k)}_{N-n_k+1} = k_{\rm B}T\ln\frac{Z^{(k)}_{N-n_k}}{Z^{(k)}_{N-n_k+1}}, \tag{B.10}$$

which corresponds exactly with the expression $\mu^{(k)}_{N-n_k+1} = F^{(k)}_{N-n_k+1} - F^{(k)}_{N-n_k}$ with $F^{(k)}_N = -k_{\rm B}T\ln Z^{(k)}_N$ the Helmholtz free energy of N particles. Thus, equation (B.7) can be written as

$$\frac{Z^{(k)}_{N-n_k}}{Z^{(k)}_N} = {\rm e}^{\beta\mu_N}\,{\rm e}^{\beta\mu_{N-1}}\cdots{\rm e}^{\beta\mu_{N-n_k+1}}. \tag{B.11}$$

In the thermodynamic limit $N \to \infty$, we can write ${\rm e}^{\beta\mu_N} = \cdots = {\rm e}^{\beta\mu_{N-n_k+1}} \approx {\rm e}^{\beta\mu}$, and therefore

$$Z_N = Z^{(k)}_N\sum_{n_k=0}^{j}{\rm e}^{-\beta n_k(\epsilon_k-\mu)} = Z^{(k)}_N\frac{{\rm e}^{-\beta(\epsilon_k-\mu)(j+1)}-1}{{\rm e}^{-\beta(\epsilon_k-\mu)}-1}. \tag{B.12}$$

Consequently, by the use of (B.6) and after some algebra, we finally get

$$\langle n_k\rangle_j = \frac{1}{{\rm e}^{\beta(\epsilon_k-\mu)}-1} - \frac{j+1}{{\rm e}^{\beta(\epsilon_k-\mu)(j+1)}-1}. \tag{B.13}$$

The generalization presented here shows how the chemical potential μ emerges from the particle conservation requirement for any order of the IQS.

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