Dimensional transitions in thermodynamic properties of ideal Maxwell–Boltzmann gases

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Received 18 November 2014
Accepted for publication 17 February 2015
Published 27 March 2015

Abstract
An ideal Maxwell–Boltzmann gas confined in various rectangular nanodomains is considered under quantum size effects. Thermodynamic quantities are calculated from their relations with the partition function, which consists of triple infinite summations over momentum states in each direction. To obtain analytical expressions, summations are converted to integrals for macrosystems by a continuum approximation, which fails at the nanoscale. To avoid both the numerical calculation of summations and the failure of their integral approximations at the nanoscale, a method which gives an analytical expression for a single particle partition function (SPPF) is proposed. It is shown that a dimensional transition in momentum space occurs at a certain magnitude of confinement. Therefore, to represent the SPPF by lower-dimensional analytical expressions becomes possible, rather than numerical calculation of summations. Considering rectangular domains with different aspect ratios, a comparison of the results of derived expressions with those of summation forms of the SPPF is made. It is shown that analytical expressions for the SPPF give very precise results with maximum relative errors of around 1%, 2% and 3% at exactly the transition point for single, double and triple transitions, respectively. Based on dimensional transitions, expressions for free energy, entropy, internal energy, chemical potential, heat capacity and pressure are given analytically valid for any scale.

Keywords: quantum size effects, nano thermodynamics, confinement effects, dimensional transitions

1. Introduction
Advancements in nanoscience and nanotechnologies have increased the importance of studies related to the nanoscale properties of matter in recent years. In the presence of quantum confinement effects, thermodynamic properties of systems differ from those of macro scale. Due to the wave character of particles, quantum size effects (QSEs) appear in nanoscale systems as a consequence of quantum confinement.

Thermodynamics under QSEs has become an active research area and many novel aspects such as quantum boundary layers, anisotropic gas pressure, loss of additivity in extensive properties, quantum forces and gas diffusion due to size difference in both Maxwell–Boltzmann (MB) and quantum (Fermi–Dirac and Bose–Einstein) gases have been studied in literature [1–17]. Even though size and shape dependences in thermodynamics of nanosystems have been recently studied, dimensional transitions due to QSEs in thermodynamic quantities and transition conditions have not been examined in the literature.

Thermodynamic state functions are represented by summations from their definitions in statistical mechanics [18, 19]. In thermodynamics of macrosystems, it is common to replace summations by integrals to decrease the calculation burden and to obtain analytical expressions. Conversion of summations to integrals is done by using the continuum approximation and density of states concept, under the assumption that discrete energy levels are so close to each other that they can be considered as continuous variables. The continuum approximation works extremely well on the macroscale. However, on the nanoscale, ascending divergence appears between summations and their integral approximations [7, 19].
At mesoscales, instead of integral approximations, Weyl’s conjecture or the first two terms of the Poisson summation formula (PSF) are used in literature [2–5, 7–12, 19]. On the other hand, when characteristic sizes of the domain are smaller than half of the most probable de Broglie wavelengths of particles, even these approximations considered in the literature become insufficient. In the case of such strong confinements, one should either consider the numerical calculation of exact summations in thermodynamic properties or look for a better solution.

In this article, a method based on dimensional transitions in momentum space due to quantum confinement is proposed for the calculation of the single particle partition function (SPPF) of MB statistics. It is seen that the method leads to analytical expressions which give nearly the same results as exact summations. Then, from the analytical expressions of the SPPF, thermodynamic quantities such as free energy, entropy, internal energy, heat capacity and pressure are represented thermodynamic behavior of strongly confined systems without the need to calculate relevant triple summations.

2. Determination of dimensional transition point in momentum space

For a high temperature and/or low density ideal gas, the SPPF is given in triple summation form by using MB statistics as follows:

\[
\zeta = \sum_{i_1} \exp\left(-\frac{\varepsilon_{i_1}}{k_B T}\right)
\]

where \(i_1 = \{1, 2, 3, \ldots\}\) are momentum state variables with subscript of \(n = \{1, 2, 3\}\) indicating directions, \(\varepsilon\) are the energy eigenvalues from the solution of the Schrödinger equation for a rectangular domain, \(k_B\) is Boltzmann’s constant and \(T\) is temperature. By defining a confinement parameter \(\alpha_i = L_{ci}/L_{ci}\) that indicates the magnitude of confinement in the \(n\)th direction, equation (1) can also be written as follows:

\[
\zeta = \prod_{n=1}^{3} \sum_{i_n=1}^{\infty} \exp\left(-\alpha_i \varepsilon_{i_n}\right)^2.
\]

In the definition of \(\alpha_i\), \(L_{ci}\) is the size of the domain in the \(n\)th direction and \(L_{ci} = h/\sqrt{8\pi \hbar k_B T}\) is half of the most probable de Broglie wavelength of particles where \(h\) is Planck’s constant and \(m\) is the mass of the particle.

Although triple summations are the exact way to calculate the SPPF, on macroscales, where \(\alpha\) values are much smaller than unity \((\alpha \ll 1)\), summations are replaced by integrals making a continuum approximation. On the other hand, when domain size in a particular direction \(L_{ci}\) is of the order of \(L_{ci}\) as on the nanoscale \((\alpha \approx 1)\), there is a considerable deviation between the results of summations and integrals. Therefore, a more precise evaluation method, such as the Poisson summation formula (PSF), is necessary to calculate summations. The PSF is written for even functions, \(f(i) = f(-i)\), as

\[
\sum_{i=1}^{\infty} f(i) = \int_{0}^{\infty} f(i) di - \frac{f(0)}{2}
\]

\[
+ \frac{\alpha}{2} \sum_{i=1}^{\infty} \int_{0}^{\infty} f(i) \cos(2\pi i) di.
\]

By evaluating each term of the PSF, the SPPF in 1D can be exactly represented as

\[
\zeta_{1D} = \sum_{i_1=1}^{\infty} \exp \left[-\left(\alpha \varepsilon_{i_1}\right)^2\right]
\]

\[
= \frac{1}{2\pi} - \frac{1}{2} + \frac{\sqrt{\pi}}{\alpha_1} \sum_{i_1=1}^{\infty} \exp \left[-\left(\frac{\pi s}{\alpha_1}\right)^2\right].
\]

The first term of the PSF is the conventional integral term, and its accuracy for macrodomains \((\alpha \rightarrow 0)\) is quite good. When domain size gets closer to \(L_{ci} \rightarrow 1\), the contribution of the second term (called the zero correction term) of the PSF becomes appreciable. The first two terms of the PSF can be called the one-dimensional (1D) representation, since they represent the contributions of whole momentum state variables, \(i_1 = \{1, \ldots, \infty\}\). The contribution of the zero correction term is studied as a QSE in literature [7, 8, 10–12, 15, 16], especially for weakly confined domains \((\alpha < 1)\), in which the contribution of the third term of the PSF is negligible. To fully represent the SPPF on any scale, even for strongly confined domains \((\alpha > 1)\), whole terms of the PSF have to be considered. However, since there is no known analytical solution for the third term which includes another infinite summation, either we have to calculate infinite summations of the SPPF numerically or find a method to give analytical expressions representing the exact behavior of the SPPF successfully. For strongly confined cases \((\alpha > 1)\), fortunately, the contribution of just the first term \((i_1 = 1)\) of the SPPF’s summation becomes extremely dominant. Therefore, it can be called the zero-dimensional (0D) representation, since it represents almost only the contribution of the ground state.

To see the success of 1D and 0D representations, variations of their analytical expressions with confinement parameter are compared with that of the exact 1D SPPF based on numerical calculation of the summation, in figure 1. The solid curve shows the variation of the exact form of the 1D SPPF (equation (4a)) with confinement parameter \(\alpha_1\). Plus signs show the results of 1D analytical representation based on the first two terms of the PSF, while cross signs demonstrate the results of 0D representation based on just the first term \((i_1 = 1)\) of the SPPF’s summation.

It is understood that the 1D analytical expression matches perfectly with the exact form of the SPPF until a certain value of \(\alpha = \alpha^*\), after which it considerably deviates. In the same manner, the 0D expression substantially deviates before \(\alpha^*\), while it perfectly matches with the solid curve after this. This certain value of \(\alpha\), which corresponds to the transition point from 1D to 0D representation, can be found as

2
\[ \alpha^* = 1.23547 \ldots \text{ by equating the 1D expression to the 0D one.} \]

Therefore, an analytical expression representing the SPPF for any values of \( \alpha \) can be given as follows:

\[
\sum_{n=1}^{\infty} \exp\left[-\left(a_n\right)^2\right] \\
\approx \left( \frac{\sqrt{\pi}}{2a_1} \right)^c \left[ \exp\left(-a_1^2\right) \right]^{1-c} \\
- \frac{c}{2} \begin{cases} 
2 & \text{for } a_1 < \alpha^* \\
\infty & \text{for } a_1 \geq \alpha^*
\end{cases}
\]

(5)

where the transition value is \( \alpha^* = 1.23547 \ldots \) This approximation gives 1% error even at the transition point. The transition represents the dimensional change (contraction/expansion) of momentum space in a particular direction. Henceforward, the domain in the \( n \)th direction will be considered as free and confined for \( \alpha_n < \alpha^* \) and \( \alpha_n \geq \alpha^* \) respectively.

### 2.1. Dimensional transitions and superpositions in SPPF

By using the dimensional transition point in momentum space, we may now consider SPPFs for the domains which are confined at the nanoscale in certain directions. The 3D domain is free in all three directions, so that confinement values in all directions are smaller than \( \alpha^* \), the transition value. For the 2D domain, \( \alpha_1 \), and for the 1D domain, \( \alpha_1 \) and \( \alpha_2 \), are chosen as greater than \( \alpha^* \) respectively. The 0D domain is considered as confined in all directions, which means all of its confinement parameters are larger than \( \alpha^* \). In this sense, using equation (5), we can write analytical expressions for the SPPF of an ideal MB gas for 3D, 2D, 1D and 0D domains as

\[
\zeta_{3D} = \frac{\pi^{3/2}}{8a_1a_2a_3} \left(1 - \frac{\alpha_1}{\sqrt{\pi}}\right) \\
\times \left[ \left(1 - \frac{\alpha_2}{\sqrt{\pi}}\right) \left(1 - \frac{\alpha_3}{\sqrt{\pi}}\right) \right] \\
\zeta_{2D} = \frac{\pi}{4a_2a_3} \exp\left(-\alpha_1^2\right) \left(1 - \frac{\alpha_3}{\sqrt{\pi}}\right) \\
\times \left[ \left(1 - \frac{\alpha_2}{\sqrt{\pi}}\right) \{a_1\} > \alpha^* \right]
\]

(6)

\[
\zeta_{1D} = \frac{\sqrt{\pi}}{2a_3} \exp\left(-\alpha_1^2 - \alpha_2^2\right) \\
\times \left[ \left(1 - \frac{\alpha_3}{\sqrt{\pi}}\right) \{a_1, a_2\} > \alpha^* \right] \\
\zeta_{0D} = \exp\left(-\alpha_1^2 - \alpha_2^2 - \alpha_3^2\right).
\]

\[
\{a_1, a_2, a_3\} > \alpha^*
\]

Although the dimensional transition point has a unique value, by engaging the sizes of the domains to each other in each direction, it is possible to obtain different transition points correspond to different dimensional transitions for anisometric domains. In order to engage sizes to each other, we may define constant aspect ratios between sizes in three directions. Therefore, we can change the size of the domain by changing only one of the sizes, which is chosen here as \( L_1 \), while preserving its shape. The aspect ratio between sizes in the first and second directions is denoted by \( n_2 = L_1/L_2 \) and in the second and third directions by \( n_3 = L_2/L_3 \). In this case, the confinement parameters of the second and third directions can be written in terms of the confinement parameter in the first direction as \( \alpha_2 = \alpha_1n_2 \) and \( \alpha_3 = \alpha_1n_2n_3 \) respectively. In figure 2, relative errors (\( R_{\zeta_n} \)) of analytical expressions given by equation (6) are given for different aspect ratios.

Peaks in figure 2 correspond to the values \( \alpha^*, \alpha^*/n_2 \) and \( \alpha^*/n_2n_3 \) for transitions from 3D to 2D, 2D to 1D and 1D to 0D respectively. As seen from figure 2(a), each dimensional transition causes an error of 1% even at the transition points. If the sizes in two different directions are the same, dimensional transitions in these directions superpose, this causes 2% error, as seen in figures 2(b) and (c). Similarly, for an isometric domain, superposition of three different dimensional transitions causes 3% error (figure 2(d)).

### 3. Dimensional transitions in thermodynamic properties

The free energy of an ideal MB gas consisting of \( N \) particles is simply given by \( F = -Nk_B T \ln(\zeta/N) + 1 \) [18, 19]. Dimensionless free energy can be defined as \( \tilde{F} = F/Nk_B T \). Then, we can obtain analytical expressions for dimensionless
free energies of a MB gas for 3D, 2D, 1D and 0D cases by considering dimensional transitions

\[
\tilde{F}_{3D} = -\ln\left(\frac{\pi^{3/2}}{8N\alpha_1\alpha_2\alpha_3}\right) - 1 + \ln(q_1 + 1) + \ln(q_2 + 1) + \ln(q_3 + 1)
\]

\[
\tilde{F}_{2D} = -\ln\left(\frac{\pi}{4\alpha_1\alpha_2}\right) - 1 + \alpha_1^2 + \alpha_2^2 + \ln(q_1 + 1) + \ln(q_2 + 1), \{\alpha_1, \alpha_2\} > \alpha^*
\]

\[
\tilde{F}_{1D} = -\ln\left(\frac{\sqrt{\pi}}{2\alpha_1}\right) - 1 + \alpha_1^2 + \alpha_2^2 + \ln(q_1 + 1), \{\alpha_1, \alpha_2\} > \alpha^*
\]

\[
\tilde{F}_{0D} = -\ln\left(\frac{1}{N}\right) - 1 + \alpha_1^2 + \alpha_2^2 + \alpha_3^2, \{\alpha_1, \alpha_2, \alpha_3\} > \alpha^*
\]

respectively, where \(q_n = \alpha_n/(\sqrt{\pi} - \alpha_n)\) with \(n = \{1, 2, 3\}\) as the direction index. It is clear that \(\alpha_n\) is always smaller than \(\sqrt{\pi}\) since \(q_n\) is necessary only for the case of \(\alpha_n < \alpha^*\). Note that we can use MB statistics if and only if the \(1 \ll N \ll 1/\alpha_1\alpha_2\alpha_3\) condition holds. This condition holds even if one of the confinement parameters \(\alpha_n\) is a little smaller than unity, because in this condition the system contains enough particles to carry out statistics \((N \gg 1)\) and the interparticle distance is much greater than \(L_c\) to use MB statistics. Since the 0D case does not satisfy this condition, one could not use MB statistics for 0D conditions. Nevertheless, in this article, we also give 0D expressions of thermodynamic properties, just for mathematical intuition. Therefore, it is worth noting that thermodynamic expressions given for 0D do not represent a meaningful physical situation, since the condition of \(N \ll 1/\alpha_1\alpha_2\alpha_3\) leads to \(N < 1\) for the 0D condition.

From free energies, we can derive all other thermodynamic quantities such as entropy, internal energy, chemical potential, heat capacity at constant sizes and pressure as

\[
S = -\left(\frac{\partial F}{\partial T}\right)_{N,V} \quad U = F + TS
\]

\[
\mu = \left(\frac{\partial F}{\partial N}\right)_{T,V} \quad C = \left(\frac{\partial U}{\partial T}\right)_{L_i,L_2,L_3}
\]

\[
P_m = \frac{N}{V} k_B T \frac{L_n}{\epsilon}\left(\frac{\partial \epsilon}{\partial L_n}\right)_T
\]

Thermodynamic properties can be written in their dimensionless forms as \(\bar{S} = S/Nk_B\), \(\bar{U} = U/Nk_B T\), \(\bar{\mu} = \mu/Nk_B T\), \(\bar{C} = C/Nk_B\) and \(\bar{P}_m = P_m/V/Nk_B T\), where \(V = L_1L_2L_3\). Even though pressure is a scalar quantity on the macroscale, it becomes a tensorial quantity with zero off-
diagonal elements for anisometric domains. In other words, \( P_1 \neq P_2 \neq P_3 \) for an anisometric rectangular domain where \( L_1 \neq L_2 \neq L_3 \).

Dimensionless entropy, internal energy, chemical potential, heat capacity and pressure expressions for MB gases are derived by considering dimensional transitions and are given in Table 1.

It is easy to check that all expressions derived in this article go to their classical expressions when the confinement parameters \( \alpha_n \) go to zero. Therefore, they are valid not only for nanosystems but also for meso- and macrosystems. They are generic in this sense, within the error limit at dimensional transition points.

Note for heat capacity that, instead of referring to constant volume, we refer here to constant sizes, which means sizes in each direction \( L_1, L_2 \) and \( L_3 \) are constant. Otherwise, combination of different \( L_n \) values can also correspond to the same volume, although they correspond to different heat capacity values, different from that on the macroscale.

### Table 1. Thermodynamic properties of Maxwell–Boltzmann gases in various dimensional conditions

| Property         | Expression                                                                 |
|------------------|-----------------------------------------------------------------------------|
| Entropy          | \(
\tilde{S}_{1D} = \frac{s}{2} + \ln \left( \frac{s^{3/2}}{N_{\text{MB}}^*} \right) - \ln(q_1 + 1) - \ln(q_2 + 1) - \ln(q_3 + 1)
\)
\( + \frac{1}{s} \left( q_1 + q_2 + q_3 \right) \)
\( \tilde{S}_{2D} = 2 + \ln \left( \frac{s}{N_{\text{MB}}^*} \right) - \ln(q_2 + 1) - \ln(q_3 + 1) + \frac{1}{s} \left( q_2 + q_3 \right) \) \( \left( \alpha_1 \right) \)
\( > a^* \)
\( \tilde{S}_{3D} = \frac{s}{2} + \ln \left( \frac{s}{N_{\text{MB}}^*} \right) - \ln(q_3 + 1) + \frac{1}{s} \left( q_3 \right), \ \left\{ a_1, a_2 \right\} > a^* \)
\( \tilde{S}_{4D} = 1 + \ln \left( \frac{s}{2} \right), \ \left\{ a_1, a_2, a_3 \right\} > a^* \)
| Internal energy  | \( \tilde{U}_{1D} = \frac{s}{2} + \frac{1}{s} \left( q_1 + q_2 + q_3 \right), \ \left\{ a_1 \right\} > a^* \)
\( \tilde{U}_{2D} = 1 + a_1^2 + \frac{1}{s} \left( q_2 + q_3 \right), \ \left\{ a_1 \right\} > a^* \)
\( \tilde{U}_{3D} = \frac{s}{2} + a_1^2 + a_2^2 + \frac{1}{s} \left( q_3 \right), \ \left\{ a_1, a_2 \right\} > a^* \)
\( \tilde{U}_{4D} = a_1^2 + a_2^2 + a_3^2, \ \left\{ a_1, a_2, a_3 \right\} > a^* \)
| Chemical potential| \( \tilde{\mu}_{1D} = -\ln \left( \frac{s^{3/2}}{N_{\text{MB}}^*} \right) + \ln(q_1 + 1) + \ln(q_2 + 1) + \ln(q_3 + 1) \)
\( \tilde{\mu}_{2D} = -\ln \left( \frac{s}{N_{\text{MB}}^*} \right) + a_1^2 + \ln(q_2 + 1) + \ln(q_3 + 1), \ \left\{ a_1 \right\} \)
\( > a^* \)
\( \tilde{\mu}_{3D} = -\ln \left( \frac{s}{2N_{\text{MB}}^*} \right) + a_1^2 + a_2^2 + \ln(q_3 + 1), \ \left\{ a_1, a_2 \right\} > a^* \)
\( \tilde{\mu}_{4D} = -\ln \left( \frac{s}{4N_{\text{MB}}^*} \right) + a_1^2 + a_2^2 + a_3^2, \ \left\{ a_1, a_2, a_3 \right\} > a^* \)
| Heat capacity at constant sizes \( \left\{ L_1, L_2, L_3 \right\} \) | \( \tilde{C}_{1D} = \frac{s}{2} + \frac{1}{s} \left[ q_1 (1 - q_1) + q_2 (1 - q_2) + q_3 (1 - q_3) \right], \ \left\{ a_1 \right\} > a^* \)
\( \tilde{C}_{2D} = 1 + \frac{1}{s} \left[ q_1 (1 - q_1) + q_2 (1 - q_2) \right], \ \left\{ a_1 \right\} > a^* \)
\( \tilde{C}_{3D} = \frac{s}{2} + \frac{1}{s} \left[ q_1 (1 - q_1) \right], \ \left\{ a_1 \right\} > a^* \)
\( \tilde{C}_{4D} = 0, \ \left\{ a_1, a_2, a_3 \right\} > a^* \)
| Pressure         | \( \tilde{P}_{1D} = (q_1 + 1), \ \tilde{P}_{2D} = (q_2 + 1), \ \tilde{P}_{3D} = (q_3 + 1) \)
\( \tilde{P}_{1D} = 2a_1^2, \ \tilde{P}_{2D} = 2a_2^2, \ \tilde{P}_{3D} = (q_3 + 1), \ \left\{ a_1 \right\} > a^* \)
\( \tilde{P}_{1D} = 2a_1^2, \ \tilde{P}_{2D} = 2a_1^2, \ \tilde{P}_{3D} = (q_3 + 1), \ \left\{ a_1, a_2 \right\} > a^* \)
\( \tilde{P}_{1D} = 2a_1^2, \ \tilde{P}_{2D} = 2a_1^2, \ \tilde{P}_{3D} = 2a_1^2, \ \left\{ a_1, a_2, a_3 \right\} > a^* \)
Unlike the scalar behavior of pressure on the macroscopic scale, quantum confinement in one direction brings a positive contribution to the pressure in this direction, which makes the pressure a tensorial quantity on the nanoscale. Besides, when the confinement in a particular direction is larger than $\alpha^*$, pressure in that direction grows quadratically.

4. Conclusion

By introducing dimensional transitions of thermodynamic properties for MB gases, it is shown that it is possible to obtain analytical expressions within a negligible error to represent the exact forms of thermodynamic properties based on triple infinite summations. In this regard, analytical results are obtained for the SPPF, free energy, entropy, internal energy, chemical potential, heat capacity and pressure, for 3D, 2D, 1D and 0D cases. It is seen that thermodynamic quantities have a strict size dependence, which cause dimensional contraction in free momentum space for nanosystems.

The critical value of the confinement parameter is found as $\alpha^* = 1.23547$. When the confinement parameter in a particular direction becomes greater than the critical value of transition, the momentum component in this direction can only take its ground-state value. Then, the degree of freedom of momentum space decreases by one unit. Therefore, depending on the values of the confinement parameter in each direction, the degree of freedom takes the values of 3,2,1 and 0. By considering this dimensional transition model, it is possible to express the thermodynamic properties analytically with 1% error for any confined system.

As expected, classical expressions can be recovered in case of the absence of confinement ($\alpha_n \to 0$). Dimensional transitions in ideal Fermi and Bose gases are under consideration. Thermodynamic properties of strongly confined ideal Fermi and Bose gases in nanodomains will then be analytically obtained for any scale.

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