

Thermodynamics of Particle Systems in the Presence of External Macroscopic Fields

I. Classical Case

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Received March 30, 1972

Abstract. We study the grand partition function of a system of identical particles interacting via a superstable potential in the presence of an external field depending on a scale factor. We discuss the case when the scale factor increases to infinity (macroscopic limit for the external potential) and we prove rigorously a link between the so obtained pressure and the usual one (barometric formula).

1. Introduction

Many efforts have been devoted to the discussion of the thermodynamic behaviour of macroscopic systems in a rigorous statistical framework [1]. In this spirit the notion of macroscopic potential has not yet been fully considered¹. When a gas of particles is in thermodynamic equilibrium in an external potential, the external field is considered macroscopic if the following requirement is fulfilled: it is possible to divide the whole space in subregions small enough for the potential to be approximately constant in them, but large enough to consider in each region statistically independent systems.

The aim of this paper is to discuss these topics in a quantitative and rigorous way. We study the classical grand partition function of a system of identical interacting particles in an external field depending on a scale factor. The macroscopic limit for the external potential is achieved letting the scale factor increase to infinity. As a result a link is found between the so obtained pressure and the usual one (barometric formula).

We remark that the previous procedure can provide by a small external field an explicit breakdown of the symmetry of statistical systems.

* Research partially supported by the Consiglio Nazionale delle Ricerche.

¹ Recently a similar problem has been discussed for classical particles with an hard core: see [2] and [3].

It can also be useful for discussing the thermodynamic limit with not rigid external walls. Finally sometimes it is possible to solve exact models in an external field, and then come back to the pressure in the thermodynamic limit with rigid walls [4, 5].

In order that this macroscopic limit exist the natural requirement on the mutual interaction would be the stability; but to avoid anomalous large fluctuations of the number of particles in finite regions, we shall consider the particles interacting via superstable potentials². As stressed by Ruelle this is not a real loss of generality with respect to the class of stable potentials [6].

The quantum extension of the previous results will be discussed in a subsequent paper.

2. Notations, Assumptions and Results

We consider a system of identical particles interacting via a potential U and moving in an external potential V . We assume

(D.1) Let $V: \mathbb{R}^v \rightarrow [0, \infty]$ such that $\exp[-V(x)]$ is a Riemann integrable function in \mathbb{R}^v . The external potential is then defined to be $V(\gamma x)$ where $\gamma \in (0, 1]$.

(D.2) Let $\Phi: [0, \infty] \rightarrow \mathbb{R}$ with Φ Lebesgue measurable. The inter-particle configuration energy is then defined to be

$$U(x_1, \dots, x_m) = \sum_{i=2}^m \sum_{j=1}^{i-1} \Phi(\|x_i - x_j\|) \quad x_1, \dots, x_m \in \mathbb{R}^v.$$

Note. For the sake of simplicity we have considered only pairwise interactions; the results we obtain hold also in the general case.

(D.3) Classical stability: there exists $B \geq 0$ such that for all m, x_1, \dots, x_m

$$U(x_1, \dots, x_m) \geq -mB$$

(D.4) Weak-tempering: there exist $K_1 \geq 0, \alpha > v, x_0 > 0$ such that

$$\Phi(x) \leq K_1 x^{-\alpha} \quad \text{for } x \geq x_0$$

It is now possible to define for our system the grand canonical partition function and the corresponding pressure as

(D.5)

$$Z_\gamma(\mu, \beta) = \sum_{n=0}^{\infty} \frac{1}{n!} \exp(\beta \mu n) \cdot \lambda^n \cdot \int_{\mathbb{R}^{vn}} dx_1 \dots dx_n \exp \left[-\beta \left(U(x_1, \dots, x_n) + \sum_{i=1}^n V(\gamma x_i) \right) \right]. \quad (2.1)$$

² Our results hold also for some not necessary superstable interactions, for instance for free particles and in general for non negative potentials.

where $\lambda \exp(\beta\mu)$ is the activity.

$$P_\gamma(\mu, \beta) = \gamma^v \beta^{-1} \ln Z_\gamma(\mu, \beta) \tag{2.2}$$

We shall also use the pressure in absence of external fields both for finite and infinite volumes

(D.6)

$$P(\mu, \beta, A) = \beta^{-1} |A|^{-1} \ln \left\{ \sum_{n=0}^{\infty} (n!)^{-1} \lambda^n \exp(\beta\mu n) \cdot \int_{A^n} dx_1 \dots dx_n \exp[-\beta U(x_1, \dots, x_n)] \right\}. \tag{2.3}$$

When the volume of the region A increases to infinity in the Fisher sense [1] we define

$$P(\mu, \beta) = \lim_{|A| \rightarrow \infty} P(\mu, \beta, A) \tag{2.4}$$

In the sequel we shall also need the two following further assumptions on the interaction

(D.7) Superstability: let $\Phi = \Phi' + \Phi''$ where Φ' is stable and Φ'' is a continuous non negative function with $\Phi''(0) > 0$.

(D.8) Lower regularity: there exist $K_2 \geq 0, \alpha > \nu$ such that

$$\Phi(x) \geq -K_2 x^{-\alpha} \quad \text{for } x \geq 0.$$

The main result of this paper is the following theorem, which proves the existence of the thermodynamic limit of the pressure defined in (D.5) and gives an explicit link with the rigid walls pressure of Eq.(2.4).

Theorem 1. *Let Φ satisfy (D.2), (D.4), (D.7) and (D.8) and V satisfy (D.1) then*

$$\lim_{\gamma \rightarrow 0} \beta^{-1} \gamma^v \ln Z_\gamma(\mu, \beta) = \int_{\mathbb{R}^v} dx P(\mu - V(x), \beta)$$

In the sequel we shall use the sets of cubes

$$\Gamma_l(r) = \{x \in \mathbb{R}^v : (r^i - \frac{1}{2}) l \leq x^i < (r^i + \frac{1}{2}) l \quad i = 1, \dots, v\}, \tag{2.5}$$

$$\Gamma_{l,R}(r) = \{x \in \mathbb{R}^v : (r^i - \frac{1}{2}) l + R \leq x^i < (r^i + \frac{1}{2}) l - R \quad i = 1, \dots, v\} \tag{2.6}$$

where $l \in \mathbb{Z}^+$ and $r \in \mathbb{Z}^v$ and $0 < R < \frac{l}{2}$.

We shall also use the following notation

$$|x| = \max_{1 \leq i \leq v} |x^i| \quad x \in \mathbb{R}^v, \tag{2.7}$$

$$\mathcal{I}(a) = \text{integer part of } a, \quad a \in \mathbb{R}. \tag{2.8}$$

3. Macroscopic Limit

In this section we give the proof of Theorem 1. First we prove two lemmata which give a lower and an upper bound.

Lemma 1 (*Lower bound*). *Let Φ satisfy (D.2), (D.3), (D.4), then*

$$\liminf_{\gamma \rightarrow 0} \beta^{-1} \gamma^v \ln Z_\gamma(\mu, \beta) \geq \int_{\mathbb{R}^v} dx \mathbb{P}(\mu - V(x), \beta).$$

Proof. We now sketch the main line of the proof. We follow the physical ideas discussed in the introduction. We divide the space in cells separated by corridors. For evaluating the interaction between different cells we limit the density in each region. Then we perform the macroscopic limit, so that the external potential becomes constant in each cell. Successively we prove that there exist a suitable way to go to infinity for the cutoff on the density and the sizes of the cells and the corridors so that the mutual interaction is eliminated and the pressure is reconstructed.

We consider in \mathbb{R}^v the two sets of cubes $\{I_l(r)\}$ and $\{I_{l,\mathbf{R}}(r)\}$ defined by Eqs. (2.5), (2.6) with the condition

$$\mathbf{R} \geq x_0 \tag{3.1}$$

and x_0 is defined in (D.4). The set $\{I_l(r)\}$ determines a partition of \mathbb{R}^v , the cubes $I_{l,\mathbf{R}}(r)$ are obtained from the cubes $I_l(r)$ subtracting a corridor depending on \mathbf{R} . By condition (3.1) particles belonging to different cubes of the set $\{I_{l,\mathbf{R}}(r)\}$ interact via the asymptotic form of the potential. If less than $M + 1$ particles are in each of the cubes $I_{l,\mathbf{R}}(r)$ an upper bound, $W(I_{l,\mathbf{R}}, M)$, can be easily given for the interaction of a cube with all the others:

$$W(I_{l,\mathbf{R}}, M) = K_1 M^2 \mathbf{R}^{-\alpha} \sum_{\substack{r \in \mathbb{Z}^v \\ r \neq 0}} |r|^{-\alpha} = KM^2 \mathbf{R}^{-\alpha} \tag{3.2}$$

We obtain a lower bound for $Z_\gamma(\mu, \beta)$ if we restrict the domain of integration to the region $\bigcup_{r \in \mathbb{Z}^v} I_{l,\mathbf{R}}(r) \subset \mathbb{R}^v$ and further we impose that in each cube no more than M particles are present. We have, using Eq. (3.2)

$$Z_\gamma(\mu, \beta) \geq \prod_{r \in \mathbb{Z}^v} \left[1 + \exp(-\beta KM^2 \mathbf{R}^{-\alpha}) \sum_{n=1}^M (n!)^{-1} \lambda^n \right. \\ \left. \cdot \exp\{\beta n[\mu - V(r, \mathbf{l}, \gamma)]\} \cdot \int_{[I_{l,\mathbf{R}}(r)]^n} dx_1 \dots dx_n \exp\{-\beta U(x_1, \dots, x_n)\} \right]. \tag{3.3}$$

where

$$\exp[-V(r, \mathbf{l}, \gamma)] = \inf_{x \in I_l(r)} \exp[-V(\gamma x)]. \tag{3.4}$$

Using the Hypothesis (D.1) that $\exp[-V(x)]$ is a Riemann integrable function we have

$$\liminf_{\gamma \rightarrow 0} \gamma^v \ln Z_\gamma(\mu, \beta) \geq \int_{\mathbb{R}^v} dy |I_\gamma(0)|^{-1} \ln \left[1 + \exp(-\beta K M^2 \mathbf{R}^{-\alpha}) \right. \\ \left. \cdot \sum_{n=1}^M (n!)^{-1} \lambda^n \exp[+\beta n\{\mu - V(y)\}] \cdot \int_{[I, \mathbf{r}(0)]^n} dx_1 \dots dx_n \exp\{-\beta U(x_1, \dots, x_n)\} \right]. \quad (3.5)$$

We now perform in a suitable way the limit of Eq. (3.5) for $M \rightarrow \infty$, $I \rightarrow \infty$, $\mathbf{R} \rightarrow \infty$. We restrict the integral in Eq. (3.5) to a compact region $\mathcal{D} \subset \mathbb{R}^v$; we will see later that the bound is continuous in the volume $|\mathcal{D}|$ so that the good inequality will be reconstructed. If we use the relation

$$\ln(1 + ab) \geq \ln a + \ln(1 + b), \quad 0 < a \leq 1, \quad b \geq 0$$

$$1 \geq a = \exp(-\beta K M^2 \mathbf{R}^{-\alpha})$$

$$b = \Sigma (n!)^{-1} \lambda^n \exp[\beta n(\mu - V(y))] \int_{[I, \mathbf{r}(0)]^n} dx_1 \dots dx_n \exp[-\beta U(x_1, \dots, x_n)].$$

Eq. (3.5) becomes

$$\liminf_{\gamma \rightarrow 0} \gamma^v \ln Z_\gamma(\mu, \beta) \geq -|\mathcal{D}| |I_\gamma(0)| K \beta M^2 \mathbf{R}^{-\alpha} \\ + \int_{\mathcal{D}} dy |I_\gamma(0)|^{-1} \ln \left\{ \sum_{n=0}^M (n!)^{-1} \lambda^n \exp[\beta n(\mu - V(y))] \right. \\ \left. \cdot \int_{[I, \mathbf{r}(0)]^n} dx_1 \dots dx_n \exp[-\beta U(x_1, \dots, x_n)] \right\} \geq -|\mathcal{D}| |I_\gamma(0)|^{-1} K \beta M^2 \mathbf{R}^{-\alpha} \\ + \beta |I_\gamma(0)|^{-1} \cdot |I, \mathbf{R}(0)| \cdot \int_{\mathcal{D}} dy P[\mu - V(y), \beta, I, \mathbf{R}(0)] \\ - |I_\gamma(0)|^{-1} |\mathcal{D}| \ln \left\{ 1 + \sum_{n=M+1}^{\infty} (n!)^{-1} \lambda^n \right. \\ \left. \cdot \exp(\beta \mu n) \int_{[I, \mathbf{r}(0)]^n} dx_1 \dots dx_n \exp[-\beta U(x_1, \dots, x_n)] \right\}. \quad (3.6)$$

In deriving the second inequality in Eq. (3.6) we used the relation

$$\ln(a - b) \geq \ln a - \ln(1 + b), \quad b \geq 0, \quad a \geq b + 1,$$

$$b = \sum_{n=M+1}^{\infty} (n!)^{-1} \lambda^n \exp[\beta n(\mu - V(y))] \\ \cdot \int_{[I, \mathbf{r}(0)]^n} dx_1 \dots dx_n \exp[-\beta U(x_1, \dots, x_n)].$$

$$a = \sum_{n=0}^{\infty} (n!)^{-1} \lambda^n \exp[\beta n(\mu - V(y))] \\ \cdot \int_{[I, \mathbf{r}(0)]^n} dx_1 \dots dx_n \exp[-\beta U(x_1, \dots, x_n)].$$

Using the stability we can now bound from below the third term in the r.h.s. of Eq. (3.6), and then perform the limit for $M, \mathbf{l}, \mathbf{R}$ going to infinity in a way that $M \gg \mathbf{l} \gg \mathbf{R}$. Namely chosen η verifying

$$0 < \eta < (\alpha - \nu) \nu^{-1} (\nu + 2a) < (2\nu)^{-1}$$

we let M go to infinity and for every M larger than a fixed M_0 we pose

$$\mathbf{l} = M^{\nu^{-1} - \eta}, \quad \mathbf{R} = M^{\nu^{-1} - 2\eta}.$$

It is easy to find that

$$\liminf_{\gamma \rightarrow 0} \beta^{-1} \gamma^\nu \ln Z_\gamma(\mu, \beta) \geq \int_{\mathcal{D}} dy \mathbf{P}(\mu - V(y), \beta) \tag{3.7}$$

We used the Lebesgue theorem noting that, by stability,

$$\begin{aligned} & \beta \mathbf{P}(\mu - V(y), \beta, \Gamma_{\mathbf{l}, \mathbf{R}}(0)) \tag{3.8} \\ & \leq |\Gamma_{\mathbf{l}, \mathbf{R}}(0)|^{-1} \ln \left\{ \sum_0^\infty (n!)^{-1} \exp[\beta n(\mu - V(y))] \lambda^n \exp(\beta Bn) |\Gamma_{\mathbf{l}, \mathbf{R}}(0)|^n \right\} \\ & = \lambda \exp(\beta B + \beta \mu) \exp[-\beta V(y)] \end{aligned}$$

which is a summable function in \mathbb{R}^ν . This circumstance allows to perform the limit in Eq. (3.7) for the region \mathcal{D} invading \mathbb{R}^ν so that the proof is completed. Q.E.D.

In order to obtain the required upper bound we use the probability estimates given by Ruelle [6]. From his results it is in fact possible to deduce in a straightforward way the following lemma³.

Lemma 2. *Let Φ satisfy (D.2), (D.7) and (D.8), then given a bounded Lebesgue measurable region A , there exist $\alpha > 0 \xi > 0$ such that for every integer $m \geq 0$*

$$\begin{aligned} & \sum_{n=0}^\infty (m!)^{-1} (n!)^{-1} \lambda^{n+m} \exp[\beta \mu(n+m)] \int_{A^m} dx_1 \dots dx_m \int_{[\mathbb{R}^\nu \setminus A]^n} dy_1 \dots dy_n \\ & \cdot \exp \left[-\beta U(x_1, \dots, x_m, y_1, \dots, y_n) - \beta \sum_{i=1}^m V(\gamma x_i) - \beta \sum_{i=1}^n V(\gamma y_i) \right] \\ & \leq (m!)^{-1} \exp(-\beta \tilde{V}_A m) \xi^m \exp(-\alpha m^2) Z_\gamma(\mu, \beta) \end{aligned}$$

where

$$\exp[-\beta \tilde{V}_A] = \int_A dx \exp[-\beta V(\gamma x)]$$

Lemma 3. *(Upperbound). Let Φ satisfy (D.2), (D.4), (D.7) and (D.8), then*

$$\limsup_{\gamma \rightarrow 0} \beta^{-1} \gamma^\nu \ln Z_\gamma(\mu, \beta) \leq \int_{\mathbb{R}} dx \mathbf{P}(\mu - V(x), \beta)$$

³ It follows from the proof of Theorem 0.1 Ref. [6] when we add to the usual measure of the space the weight: $\exp[-\beta V(\gamma x)]$.

Proof. We divide again the space in cells. The proof would be trivial if there is no mutual attraction. The proof is still simple if the local density is bounded (hard core); in this case in fact it is possible to estimate the attraction between different cells, which otherwise could be unbounded. In our case we divide the contributions to the grand partition function in two parts: the first with a cutoff in the local density, the second with large density. The latter contribution is controlled by means of Lemma 2.

For every integer $m > 0$ we divide the configuration space \mathbb{R}^{vm} into two disjoint regions, \mathbb{R}_M^{vm} and its complement $[\mathbb{R}_M^{vm}]^c$. In \mathbb{R}_M^{vm} less than $M + 1$ particles are present in each $\Gamma_1(r)$. In \mathbb{R}_M^{vm} a lower bound, $W(\mathbf{n}, M)$, can be given for the interaction of the particles in a cube $\Gamma_{2n+1}(r)$ with all the others; namely exists $a \geq 0$ such that

$$W(\mathbf{n}, M) = -aM^2 \mathbf{n}^{2v-\alpha} \quad (3.9)$$

where α is given in (D.8). Using Eq. (3.9) and Lemma 2 we can write

$$\begin{aligned} Z_\gamma(\mu, \beta) & \left[1 - \sum_{r \in \mathbb{Z}^v} \sum_{m=M+1}^{\infty} (m!)^{-1} \xi^m \exp(-\alpha m^2) \exp(-\beta m \tilde{V}_{r,1}) \right] \\ & \leq \sum_{n=0}^{\infty} (n!)^{-1} \exp(\beta \mu n) \int_{\mathbb{R}_M^v} dx_1 \dots dx_n \\ & \cdot \exp \left[-\beta U(x_1, \dots, x_n) - \beta \sum_{i=1}^n V(\gamma x_i) \right]. \end{aligned} \quad (3.10)$$

where

$$\exp(-\beta \tilde{V}_{r,1}) = \int_{\Gamma_1(r)} dx \exp[-\beta V(\gamma x)] \quad (3.11)$$

We define

$$q(\gamma) = q_0 \gamma^{-1}, \quad q_0 > 3. \quad (3.12)$$

We can then write

$$\begin{aligned} Z_\gamma(\mu, \beta) & \left[1 - \sum_{r \in \mathbb{Z}^v} \sum_{m=M+1}^{\infty} (m!)^{-1} \xi^m \exp(-\alpha m^2) \exp(-\beta \tilde{V}_{r,1} m) \right] \\ & \leq \prod_{|r| \leq \mathcal{J} \left(1 + \frac{q(\gamma)}{2n+1} \right)} \left\{ 1 + \exp(\beta a M^2 \mathbf{n}^{2v-\alpha}) \sum_{N=1}^{\infty} (N!)^{-1} \exp[\beta N(\mu - V_{r,2n+1})] \right. \\ & \cdot \int_{[\Gamma_{2n+1}(r)]^N} dx_1 \dots dx_N \exp[-\beta U(x_1, \dots, x_N)] \left. \right\} \\ & \cdot \exp \left\{ \exp[\beta(\mu + B)] \cdot \gamma^{-v} \int_{|y| \geq q_0} dy \exp[-\beta V(\gamma y)] \right\} \end{aligned} \quad (3.13)$$

where B is defined in (D.3) and

$$\exp(-V_{r,2n+1}) = \sup_{x \in \Gamma_{r,2n+1}} \exp[-V(\gamma x)]$$

We will choose in Eq. (3.13) the value of M in correspondence to each value of γ . There exist \tilde{b} such that for every $M \in \mathbb{Z}^+$ and $\gamma \in (0, 1]$ the following is true

$$\sum_{r \in \mathbb{Z}^v} \sum_{m=M+1}^{\infty} (m!)^{-1} \xi^m \exp[-\alpha m^2] \exp[-\beta m \tilde{V}_{r,1}] < \tilde{b} \xi^M \gamma^{-v} \exp(M - M \ln M).$$

Then $M(\gamma)$ is chosen to be

$$M(\gamma) = \mathcal{J}(vc \ln \gamma^{-1} + b), \tag{3.14}$$

$$b = 1 + c \cdot \exp(1), \quad c = (\xi + 1)(\tilde{b} + 1) \exp(1).$$

With this choice of $M(\gamma)$ it can be verified that the l.h.s. of Eq. (3.13) is always positive. Therefore we have

$$\begin{aligned} & \gamma^v \ln Z_\gamma(\mu, \beta) + \gamma^v \ln \left[1 - \sum_{r \in \mathbb{Z}^v} \sum_{m=M+1}^{\infty} (m!)^{-1} \xi^m \exp(-\alpha m^2) \exp(-\beta \tilde{V}_{r,1} m) \right] \\ & \leq \exp[\beta(B + \mu)] \cdot \int_{|y| \geq q_0} dy \exp[-\beta V(y)] \tag{3.15} \\ & + \gamma^v \sum_{z \in \mathbb{Z}^v} \left\{ \beta a M^2 M^{2v-\alpha} + \ln \sum_{N=0}^{\infty} (N!)^{-1} \exp[\beta N(\mu - V_{r,2n+1})] \right. \\ & \quad \left. \int_{[I_{2n+1}(r)]^v} dx_1 \dots dx_N \exp[-\beta U(x_1, \dots, x_N)] \right\}. \end{aligned}$$

We used the inequality

$$\begin{aligned} \ln(1 + uv) & \leq \ln(1 + v) + \ln(u), \quad u \geq 1, \quad v \geq 0, \\ u & = \exp(\beta a M^2 n^{2v-\alpha}) \\ v & = \sum_{N=1}^{\infty} (N!)^{-1} \exp[\beta N(\mu - V_{r,2n+1})] \\ & \quad \int_{[I_{2n+1}(r)]^v} dx_1 \dots dx_N \exp[-\beta U(x_1, \dots, x_N)]. \end{aligned}$$

In the limit $\gamma \rightarrow 0$ and $n \rightarrow \infty$

$$n = \mathcal{J}(\gamma^{\varepsilon-1}), \quad 0 < \varepsilon < 1.$$

Eq. (3.15) becomes

$$\begin{aligned} \limsup_{\gamma \rightarrow 0} \gamma^v \ln Z_\gamma(\mu, \beta) & \leq \beta \int_{|y| \leq q_0} P(\mu - V(y), \beta) dy \\ & + \exp[\beta(\mu + B)] \int_{|y| \geq q_0} dy \exp[-\beta V(y)]. \end{aligned}$$

By Eq. (3.8), using the Lebesgue theorem when $q_0 \rightarrow \infty$, the Lemma is proved. Q.E.D.

By Lemmata 1 and 3 the following theorem is proved.

Theorem 1. *Let Φ satisfy (D.2), (D.4), (D.7) and (D.8) and V satisfy (D.1) then*

$$\lim_{\gamma \rightarrow 0} \beta^{-1} \gamma^v \ln Z_\gamma(\mu, \beta) = \int_{\mathbb{R}^1} dx P(\mu - V(x), \beta).$$

Acknowledgments. We are indebted to G. Gallavotti for many helpful suggestions and discussions.

Note added in proof. After this paper was sent to the review, it appeared a note by K. Millard, a statistical Mechanical approach to the Problem of a Fluid in an External Field, Journ. Math. Phys. **13**, 222 (1972), in which a similar problem is discussed supposing hard core.

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