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Nonequilibrium Expressions for Entropy and Other Thermodynamic Quantities

C. G. Dufour[1]

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The entropy is written as a density series expansion involving a new kind of cumulant. These are defined as usual from the so-called reduced distribution functions. The first four terms of the series expansion of the entropy are shown to be identical to the known result. When density corrections are retained up to the μth order, the entropy is proved to obey approximately a conservation theorem. Finally, a discussion of nonequilibrium and equilibrium properties of the grand canonical ensemble is presented.

KEY WORDS: Nonequilibrium statistical mechanics; nonequilibrium expansions; entropy conservation law; fluctuation theorems; cumulants; generating functions.

1. INTRODUCTION

We consider a nonequilibrium system containing a stochastic number N of particles, i.e., we are considering a grand canonical ensemble. Let

$$P_{\{N\}}/N!$$

be the probability density for observing the following event: (a) There are exactly N particles in the system; (b) the system phase-space coordinates are to be found between the N-particle sets $\{N\}$ and $\{N\} + d\{N\}$.

The probability that there are exactly N particles in the system is obtained by integration:

$$P_N = \int d\{N\} \frac{P_{\{N\}}}{N!} \tag{1}$$

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[1] Faculté des Sciences, Université de l'Etat, Mons, Belgium.

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The Gibbs entropy S of the system is expressed in terms of the probability densities as$^{(1,2)}$

$$k^{-1}S = -\sum_{M\geqslant 0}\frac{1}{M!}\int d\{M\}P_{\{M\}}\ln[P_{\{M\}}h^{3M}] \qquad (2)$$

where k and h are the Boltzmann and Planck constants.

It is well known that S, like all other equilibrium thermodynamic quantities, has a (number) density series expansion. Out of equilibrium, we also call the density series expansion a sum of functionals integrated over an increasing number of particle coordinates.

The f functions, which are the one-, two-, three-,..., particle distribution functions (p.d.f.), called in general reduced distribution functions (r.d.f.), are defined from $P_{\{M\}}$ by

$$f_{\{M\}} = \sum_{N\geqslant M}\frac{N!}{(N-M)!}\int\frac{d\{N\}}{d\{M\}}\frac{P_{\{N\}}}{N!} = \sum_{N\geqslant 0}\frac{1}{N!}\int d\{N\}\,P_{\{N+M\}} \qquad (3)$$

$f_{\{M\}}$ is thus proportional to the Mth density power and the nonequilibrium S expansion is a density series expansion. Conversely, the $P_{\{M\}}$ may be expressed in terms of $f_{\{M+N\}}$:

$$P_{\{M\}} = \sum_{N=0}^{\infty}\frac{(-1)^N}{N!}\int f_{\{M+N\}}\,d\{N\} \qquad (4)$$

We call $S^{(u)}$ the value of S including the uth-order term, so that $S^{(u)}$ is a functional of all $\{f_{\{M\}}\}$, where $M \leqslant u$; $S^{(1)}$ is thus the Boltzmann entropy and $S^{(0)}$ is given by the contribution of the term containing h; by (2), (3), and (1), we get

$$\frac{S^{(0)}}{k} = -\sum_{M=0}^{\infty}\frac{1}{M!}\int d(1)\int d\{M-1\}\,P_{\{M\}}[\ln h^3]M = -\int d(1)f(1)\ln h^3$$
$$= -\left\{\sum_{N=0}^{\infty}NP_N\right\}\ln h^3 = -\langle N\rangle\ln h^3 = -\ln h^{3\langle N\rangle} \qquad (5)$$

where $\langle N\rangle$ is the mean number of particles contained in the system.

Several authors have written S in terms of the r.d.f.—e.g., H. S. Green$^{(3)}$ for a closed system and Nettleton and M. S. Green$^{(1)}$ for open systems; their method is, however, rather complicated. For equilibrium systems, Raveché$^{(4)}$ provided an easier demonstration, using isothermal activity derivatives of potentials of mean force and involving two kinds of cumulants. We intend to simplify this method by introducing a unique generalized kind of cumulant and to extend it to nonequilibrium cases. Isothermal activity derivatives are replaced by z derivatives, z being a parameter (put equal to 1 at the end) which is introduced by multiplying each $f_{\{M\}}$ by z^M:

$$f(\{M\}, z) \equiv z^M f_{\{M\}}$$

Moreover, if $f(\{M\}, z)$ is introduced instead of $f_{\{M\}}$ in the right-hand side of (4), the left-hand side will also depend on z and this relation will be taken as a definition for $P(\{M\}, z)$. Another way of introducing z would be by multiplying $P_{\{M\}}$ instead of $f_{\{M\}}$ by z^M; this method is closer to Raveché's since, at equilibrium, each $P_{\{M\}}$ contains the Mth power of the activity as a factor, but it is a little more tedious. We thus use the first method: This is presented in Section 2.

In Section 3 the conservation theorem for the entropy to an arbitrary density approximation is proved; the relation between this result and the H-theorem will be analyzed in a forthcoming paper.

A direct method for deriving fluctuation theorems, using the same techniques (generating function and multiple parametric derivative), is given in Section 4.

2. ENTROPY IN TERMS OF THE REDUCED DISTRIBUTION FUNCTIONS

We have to eliminate $\ln P_{\{M\}}$ from (2). We start with $\ln(P_{\{M\}}/f_{\{M\}})$. By means of (4), it may be written, introducing the z parameter,

$$\ln \frac{P(\{M\}, z)}{f(\{M\}, z)} = \ln \left[\sum_{N=0}^{\infty} \frac{(-z)^N}{N!} \int \frac{f\{M+N\}}{f\{M\}}\, d\{N\} \right] \tag{6}$$

or expanding in a Taylor series

$$\ln \frac{P(\{M\}, z)}{f(\{M\}, z)} = \sum_{n \geqslant 1} \frac{(-z)^n}{n!}\, C_n^{\{M\}} \tag{7}$$

Where the $C_n^{\{M\}}$ are the cumulants of

$$I_n^{\{M\}} \equiv \int d\{n\}\, \frac{f_{\{M+n\}}}{f_{\{M\}}} \tag{8}$$

They may be found by taking multiple derivatives of the right-hand sides of (6) and (7); the result is⁽⁵⁾

$$C_n^{\{M\}} = {\sum}'\, n!\, (-1)^{s-1}(s-1)! \bigg|_{s=\Sigma_i k_i} \prod_{i=1}^{n} \frac{1}{k_i!} \left[\frac{1}{i!}\, I_i^{\{M\}} \right]^{k_i} \tag{9}$$

where the prime indicates that the sum is over all solutions in nonnegative integers of $n = \sum_i ik_i$. Obviously, the $\{M\}$ coordinates play only a parametric role and, whatever M, the transformations are similar; in particular they are similar to usual cumulant transformations obtained by putting $\{M\} = 0$. The entropy becomes [(2), (5), (7)]:

$$\begin{aligned} \frac{S}{k} = {} & -\ln h^{3\langle N\rangle} - \sum_{M=0}^{\infty} \int d\{M\}\, \frac{P_{\{M\}}}{M!} \ln f_{\{M\}} \\ & - \sum_{M=0}^{\infty} \int d\{M\}\, \frac{P_{\{M\}}}{M!} \sum_{n=1}^{\infty} \frac{(-1)^n}{n!}\, C_n^{\{M\}} \end{aligned} \tag{10}$$

Defining

$$\sum_{\{M\}\subseteq\{m\}} (-1)^{m-M} \ln f_{\{M\}} = \omega_{\{m\}} \tag{11}$$

and applying (A.7) to each term of (10) yields

$$\begin{aligned}\frac{S}{k} = {} & -\ln[h^{3\langle N\rangle}] - \sum_{m=0}^{\infty} \frac{1}{m!} \int d\{m\}\, \omega_{\{m\}} f_{\{m\}} \\ & - \sum_{m=0}^{\infty} \frac{1}{m!} \int d\{m\} f_{\{m\}} \sum_{\{M\}\subseteq\{m\}} (-1)^{m-M} \sum_{n\geqslant 1} \frac{(-1)^n}{n!} C_n^{\{M\}}\end{aligned} \tag{12}$$

From identity (A.2) and the fact that $f_{\{0\}} = 1$ and $\omega_{\{0\}} = 0$, we obtain

$$\begin{aligned}\frac{S}{k} = {} & -\int d(1) f(1) \ln f(1) h^3 - \sum_{m=2}^{\infty} \frac{1}{m!} \int d\{m\}\, \omega_{\{m\}} f_{\{m\}} \\ & - \sum_{s=1}^{\infty} \sum_{n=1}^{s} \int \frac{d\{s\}}{d\{n\}} \frac{f\{s-n\}}{n!\,(s-n)!} \sum_{\{M\}\subseteq\{s-n\}} (-1)^{s-M} C_n^{\{M\}}\end{aligned} \tag{13}$$

The general term in the last term is also integrated over s particles since $C_n^{\{M\}}$ contains r.d.f. integrated over $\{n\}$: In each $I_i^{\{M\}}$, an integral over $\{i\}$ is hidden and $\sum_i ik_i = n$.

In fact, we have

$$C_n^{\{M\}} = \int d\{n\}\, C_{\{n\}}^{\{M\}}$$

where the $C_{\{n\}}^{\{M\}}$ are Ursell functions defined by means of $f_{\{M+n\}}/f_{\{M\}}$ instead of $f_{\{n\}}$. Thus (13) is the density series expansion we wished. As an example, it can be easily checked that the $S^{(4)}$ contribution is identical to the one calculated by Nettleton and M. S. Green,⁽¹⁾ Yvon,⁽²⁾ and Raveché.⁽⁴⁾

In this derivation, the generating function character of the method is more obvious than in Raveché's paper and the main difference between the two results is that (13) is valid whether the system is in equilibrium or not.

We can also easily obtain Green's result for closed systems⁽³⁾ with specific hypothesis of a canonical ensemble: There are exactly N_c particles in the system, i.e., all $P_{\{N\}}$ are equal zero except $P_{\{N_c\}}$. Some consequences are: r.d.f. are defined only for $M \leqslant N_c$: From (3) and (9),

$$f_{\{M\}} = \frac{1}{(N_c - M)!} \int \frac{d\{N_c\}}{d\{M\}} P_{\{N_c\}}, \qquad M \leqslant N_c$$

$$f_{\{M\}} = 0 \qquad\qquad M > N_c$$

$$C_{\{n\}}^{\{M\}} = 0, \qquad n > N_c - M$$

and in (10), n runs from 1 to $N_c - M$; thus the summation is different from zero only if $M < N_c$. As the $P_{\{M\}}$ factor is different from zero only if $M = N_c$, the last term in (10) vanishes and the canonical entropy is given by the two first terms of the right-hand side of (13).

3. GIBBS ENTROPY CONSERVATION LAW TO *u*th ORDER

As we are now interested in entropy derivatives, we drop the constant term $\ln h^{3\langle N\rangle}$, and write $S^{(u)}$ as

$$\frac{S^{(u)}}{k} = -\sum_{m=0}^{u} \frac{1}{m!}\int d\{m\}\, \omega_{\{m\}} f_{\{m\}}$$

$$-\sum_{s=1}^{u}\sum_{n=1}^{s}\int \frac{d\{s\}}{d\{n\}}\, \frac{f_{\{s-n\}}}{n!\,(s-n)!} \sum_{\{M\}\subseteq\{s-n\}} (-1)^{s-M} C_n^{\{M\}} \tag{14}$$

We prove the following theorem, expressing the entropy conservation to *u*th order:

When contributions involving more than *u* particles are neglected, the time derivative of $S^{(u)}$ is zero.

As $\partial_t P_{\{M\}}$ is simpler to handle (Liouville's theorem) than $\partial_t f_{\{M\}}$ (BBGKY equations), we shall express $S^{(u)}$ in terms of $P\{M\}$ in order to prove this theorem. Using (A.1) and (11), we obtain

$$\frac{S^{(u)}}{k} = -\sum_{m=0}^{u} \frac{1}{m!}\int d\{m\}\left[\sum_{\{M'\}\subseteq\{m\}} (-1)^{m-M'} \ln f_{\{M'\}}\right] f_{\{m\}}$$

$$-\sum_{m=0}^{u-1} \frac{1}{m!}\int d\{m\}\, f_{\{m\}} \sum_{\{M'\}\subseteq\{m\}} (-1)^{m-M'} \sum_{n=1}^{u-m} \frac{(-1)^n}{n!} C_n^{\{M'\}} \tag{15}$$

In the last term, the upper bound $u - 1$ for m could as well be u, since, if $m = u$, the upper bound for n is zero and the summation over n disappears. Here we have to go back to the $P_{\{M\}}$ formulation: By (A.5), we find

$$-\sum_{M=0}^{u} \frac{1}{M!}\int d\{M\}\, P_{\{M\}} \sum_{\{m\}\subseteq\{M\}} \sum_{\{M'\}\subseteq\{m\}} (-1)^{m-M'} \ln f_{\{M'\}}$$

$$-\sum_{m=0}^{u}\sum_{M=u+1}^{\infty}\int d\{M\}\, P_{\{M\}} \sum_{\{M'\}\subseteq\{m\}} (-1)^{m-M'} \ln f_{\{M'\}} \frac{1}{(M-m)!\, m!}$$

$$-\sum_{m=0}^{u}\sum_{M=u+1}^{\infty}\int d\{M\} P_{\{M\}} \sum_{\{M'\}\subseteq\{m\}} \frac{(-1)^{m-M'}}{(M-m)!\, m!} \sum_{n=1}^{u-m} \frac{(-1)^n}{n!} C_n^{\{M'\}}$$

$$-\sum_{M=0}^{u} \frac{1}{M!}\int d\{M\} P_{\{M\}} \sum_{\{m\}\subseteq\{M\}} \sum_{\{M'\}\subseteq\{m\}} (-1)^{m-M'}$$

$$\times\left[-\sum_{n=u-m+1}^{\infty} \frac{(-1)^n}{n!} C_n^{\{M'\}} + \sum_{n=1}^{\infty} \frac{(-1)^n}{n!} C_n^{\{M'\}}\right] \tag{16}$$

We replace the second term in the bracket by its value drawn from (7); the $\ln f_{\{M'\}}$ contribution will cancel the first term in (16), while by (A.6), the contribution of $\ln P_{\{M'\}}$ yields

$$-\sum_{M=0}^{u} \frac{1}{M!} \int d\{M\}\, P_{\{M\}} \ln P_{\{M\}} \tag{17}$$

whose time derivative, according to Liouville's equation, vanishes identically.

As time derivatives of integrals involving more than u particles may not involve less than $u + 1$ particles, the time derivative of $S^{(u)}$ gives rise only to higher order density terms: All contributions involving less than $u + 1$ particles vanish identically. This proves the theorem.

If these higher order terms involving $f_{\{u+1\}}$ (and correlation functions of the same order) are not negligible, $S^{(u+1)}$ may be considered first: This expression is now a functional of $f_{\{u+1\}}$ and its time derivative will give rise to higher order r.d.f. and perhaps could be neglected.

The theorem is thus an approximation in the sense that higher order correlation functions must always be neglected, except for $u = \infty$ (or $u = N_c$ for a canonical ensemble), in which case $S^{(u)}$ becomes the exact Gibbs entropy and the theorem becomes the usual Gibbs entropy conservation law.

4. OTHER PROPERTIES OF EQUILIBRIUM AND NONEQUILIBRIUM GENERATING FUNCTIONS USING MULTIPLE PARAMETRIC DIFFERENTIATION

We collect here two kinds of expressions which relate nonequilibrium and equilibrium thermodynamic quantities, either directly to r.d.f. or by means of cumulants.

When only Ξ, the grand canonical partition function, or its nonequilibrium equivalent, is needed, various methods can be used:

1. Functional expansion.(6)
2. Statistical arguments to find out the probability P_0 that the system is empty.(7)
3. Generating functions and multiple parametric derivatives.(8)

When Ξ derivatives are needed, only the last method is of any use; an improved version, making apparent the connection with nonequilibrium expressions, is presented here.

Finally, more direct methods are indicated.

In order to prove the following general identity involving a general generating function

$$\sum_{N=n}^{\infty} \xi^N \int \frac{d\{N\}}{d\{n\}} \frac{P\{N\}}{N!}$$
$$= \sum_{l=n}^{\infty} \frac{(\xi-1)^l}{l!} \int \frac{d\{l\}}{d\{n\}} f_{\{l\}} + \sum_{N=n}^{\infty} \sum_{l=0}^{n-1} \frac{(\xi-1)^l}{(N-l)!\,l!} \int \frac{d\{N\}}{d\{n\}} P_{\{N\}} \tag{18}$$

we only have to show that, for each M, the Mth ξ derivatives of both sides evaluated for $\xi = 1$ are equal. When $M < n$, we get an identity and when $M \geqslant n$, we again find (3).

Conversely, the right-hand side of (18) can be obtained from (3) by decomposing ξ into $1 + (\xi - 1)$.

Equating the Mth derivatives of both sides for $M \geqslant n$ and $\xi = 0$ leads to the following inversion formula:

$$M! \int \frac{d\{M\}}{d\{n\}} \frac{P_{\{M\}}}{M!} = \sum_{l=M}^{\infty} \frac{(-1)^{l-M} l!}{(l-M)!\,l!} \int \frac{d\{l\}}{d\{n\}} f_{\{l\}} \tag{19}$$

which yields the usual inversion formula (4) if $\{n\} \equiv \{M\}$. When $n = 0$, (18) reduces (1) to the useful generating function$^{(7)}$

$$\sum_{N=0}^{\infty} \xi^N P_N = \sum_{l=0}^{\infty} (\xi - 1)^l \frac{1}{l!} \int d\{l\} f_{\{l\}} \tag{20}$$

An equivalent expression is found by taking the logarithm of both sides and recalling that a logarithm of a series generates an ordinary cumulant (C_l^0) expansion (6) and (7). Those relations differentiated l times with respect to ξ yield two relations, which become the classical fluctuation theorems when equilibrium ensembles are considered$^{(8)}$:

$$\frac{\partial^l}{\partial \xi^l} \sum_{N=0}^{\infty} \xi^N P_N \bigg|_{\xi=1} = \int d\{l\} f_{\{l\}} \tag{21}$$

$$\frac{\partial^l}{\partial \xi^l} \ln\left(\sum_{N=0}^{\infty} \xi^N P_N \right) \bigg|_{\xi=1} = C^0_{\{l\}} \tag{22}$$

A shorter method consists in integrating (3) over $\{M\}$:

$$\int d\{M\} f_{\{M\}} = \sum_{N=M}^{\infty} \int d\{N\} \frac{P_{\{N\}}}{N!} \frac{N!}{(N-M)!} = \frac{\partial}{\partial \xi^M} \sum_{N=0}^{\infty} \xi^N P_N \bigg|_{\xi=1} \tag{23}$$

Equation (22) can also be derived from (23): Applying the composite function multiple derivation$^{(5)}$ to the left-hand side of (22) for $\xi = 1$ and using (23) and (9), we obtain the right-hand side of (22).

The two nonequilibrium relations leading to formulas giving Ξ at equilibrium are found from (20) and the cumulant definitions (6) and (7):

$$\Xi^{-1} = P_0 = \sum_{l=0}^{\infty} (-1)^l \frac{1}{l!} \int d\{l\} f_{\{l\}} \tag{24}$$

and

$$\Xi^{-1} = P_0 = \exp\left[\sum_{l=1}^{\infty} \frac{(-1)^l}{l!} C_l^0\right] \tag{25}$$

More directly, (24) and thus (25) come from the usual inversion relation (4).

Relation (25) is especially useful if we need an expression giving the pressure $(\ln \Xi)/\beta V$ in terms of r.d.f. Here again, cumulants appear: This is a direct consequence of the logarithmic form of the definition of the thermodynamic quantities considered (entropy and pressure).

From a mathematical point of view, we can explain why the generating function method is so widely used here: This is a very powerful method when two different series expansions may be found to be related to each other by means of a simple function; this may be a definition [e.g., (6) and (7)] or a consequence of it [18]. Multiple derivatives then yield the links between the coefficients of the two series ($C_n^{\{M\}}$ and $I_n^{\{M\}}$ or $f_{\{M\}}$ and $P_{\{M\}}$).

The right-hand sides of (6) and (7) also provide us a generating function, one relating $C_n^{\{M\}}$ and $I_n^{\{M\}}$ to each other. We can thus say that this method allowed us essentially to write down explicit expansions, in terms of reduced distribution functions, involving an increasing number of particles, for two quantities which reduce, at equilibrium, to the usual important quantities having a density series expansion: the entropy and the grand canonical partition function.

APPENDIX A

From the following obvious identity, resulting from a change of summation index

$$\sum_{m=0}^{u-d} \sum_{n=d}^{u-m} f(m, n) \equiv \sum_{s=d}^{u} \sum_{n=d}^{s} f(s - n, n) \tag{A.1}$$

we find the particular case

$$\sum_{m=0}^{\infty} \sum_{n=1}^{\infty} f(m, n) = \sum_{s=1}^{\infty} \sum_{n=1}^{s} f(s - n, n) \tag{A.2}$$

We also need the following identity:

$$\sum_{m=0}^{u} \sum_{M=u+1}^{u+n} + \sum_{M=0}^{u} \sum_{m=0}^{M} \equiv \sum_{m=0}^{u} \sum_{M=m}^{u+n} \tag{A.3}$$

which we prove by adding $\sum_{m=0}^{u}\sum_{M=u+1}^{u+n}$ to the obvious one

$$\sum_{M=0}^{u}\sum_{m=0}^{M} = \sum_{m=0}^{u}\sum_{M=m}^{u} \tag{A.4}$$

In order to prove it, we now consider the left-hand side of the following relation:

$$\sum_{m=0}^{u}\sum_{M=u+1}^{\infty}\int \frac{d\{M\}\,P_{\{M\}}}{m!\,(M-m)!}\Sigma_{(\{m\})} + \sum_{M=0}^{u}\frac{1}{M!}\int d\{M\}\,P_{\{M\}}\sum_{\{m\}\subseteq\{M\}}\Sigma_{(\{m\})}$$

$$= \sum_{m=0}^{u}\frac{1}{m!}\int d\{m\} f_{\{m\}}\Sigma_{(\{m\})} \tag{A.5}$$

where $\Sigma_{(\{m\})}$ is a functional depending of the coordinates of the group of m particles. Since the $\binom{M}{m}$ sets involving m particles give the same contribution to the integral, the left-hand side is

$$\left(\sum_{m=0}^{u}\sum_{M=u+1}^{\infty} + \sum_{M=0}^{u}\sum_{m=0}^{M}\right)\int d\{m\}\left(\int\frac{d\{M\}}{d\{m\}}P_{\{M\}}\frac{1}{m!}\right)\frac{1}{(M-m)!}\Sigma_{(\{m\})}$$

which, together with (A.3) and (3), yields the right hand side of (A.5).

A particular case is obtained by choosing $u = \infty$ and

$$\Sigma_{(\{m\})} = \sum_{\{M'\}\subseteq\{m\}}(-1)^{m-M'}l_{\{M'\}}$$

It is easy to show that the inversion formula is

$$l_{\{M'\}} = \sum_{\{m\}\subseteq\{M'\}}\Sigma_{(\{m\})}$$

and the following identity holds:

$$\sum_{\{m\}\subseteq\{M\}}\sum_{\{M'\}\subseteq\{m\}} l_{\{M'\}}(-1)^{m-M'} \equiv l_{\{M\}} \tag{A.6}$$

so that (A.5) becomes

$$\sum_{M=0}^{\infty}\frac{1}{M!}\int d\{M\}P_{\{M\}}l_{\{M\}} = \sum_{m=0}^{\infty}\frac{1}{m!}\int d\{m\} f_{\{m\}}\sum_{\{M'\}\subseteq\{m\}}(-1)^{m-M'}l_{\{M'\}} \tag{A.7}$$

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