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On a general interpretation of equilibrium entropy as a measure of randomness

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ABSTRACT. It is shown that, at the conceptual level, statistical (ensemble) entropy of an arbitrary many-body equilibrium system can be attributed a universal randomness interpretation by demonstrating the equivalent nature of an arbitrary statistical entropy variation and the corresponding one of classical ideal gas. The conceptual level consists in making use of certain well known postulates and some of their consequences that can be regarded as necessary conditions for the applicability of present-day statistical mechanics to the explanation and quantitative description of (quasi)equilibrium thermodynamic phenomena.

RESUME. On montre, au niveau du concept, que l'entropie statistique d'un système à plusieurs corps arbitraire, en équilibre, peut être dotée d'une interprétation aléatoire universelle en démontrant l'équivalence d'une variation arbitraire de cette entropie statistique avec la variation correspondante pour un gaz parfait. Se placer au niveau conceptuel revient à se servir d'un certain nombre de postulats bien connus et de leurs conséquences qui peuvent être regardés comme des conditions nécessaires d'application de la mécanique statistique actuelle à l'explication et à la description quantitative des phénomènes de quasi-équilibre thermodynamique.

1. Introduction

It is well known that physical intuition connects the concept of entropy of a given thermodynamic (macroscopic) system with the somewhat vague concept of randomness, or disorder. However "physically evident", the universality of the said interpretation of entropy is sometimes put to doubt [1] on the basis of the argument that entropy is not a property of the given physical system itself but of the thermodynamic experiments one performs on it. That is, entropy is defined in each particular case by specifying the pertinent parameters determining the thermodynamic state of our system. These parameters (and their number) are different in the various experiments, so one should avoid general statements in regard to entropy that could prove meaningless.

Such a standpoint does not seem very clear. Indeed, any thermodynamic parameter, say temperature, can be represented as a function of a specific set of parameters for a given experiment. Still, the concept of temperature is hardly made more vague (or more anthropomorphic [1] –as it is a derivative of entropy) by this fact. Evidently, the concept of equilibrium entropy and the related concept of randomness deserve a more careful analysis as well. It will be shown in this paper that a general interpretation of entropy variation as variation of randomness of microscopic state distribution is possible in a well defined sense. The generality of the said viewpoint on entropy and its variation will be of the same degree as that attributed, say to temperature : in analogy to the case of an ideal-gas thermometer which can measure, in principle, the temperature of an arbitrary thermodynamic system, the statistical description of an ideal-gas heat bath permits the attribution of a universal meaning of randomness variation to the entropy variation of an arbitrary thermodynamic system, as we shall see.

The ideology of this paper makes it possible to concentrate practically all attention on the variant of statistical mechanics employing classical mechanics as the fundamental microscopic picture underlying phenomenological (quasi)equilibrium thermodynamics. [As we are interested here in static and indefinitely slow processes and magnitudes only, the explicit specification "(quasi)equilibrium" or "(quasi)static" will sometimes be omitted hereafter.]

The consideration below will rest on well known ensemble postulates of classical statistical mechanics, the Liouville theorem, and certain consequences of these. The general randomness interpretation of entropy will emerge from the discussion of an "entropy conservation law" of a purely dynamic nature, operating in an adiabatically isolated combined system that contains as its subsystem any thermodynamic system of interest.

2. Preliminary remarks of equilibrium ensemble postulates and consistency requirements

Let us recall first the following definitions. A thermodynamic N_z-body system z ($N_z \rightarrow \infty$) is called *isolated* if it exchanges no energy of any kind with external bodies, *adiabatic* if energy exchange is due only to the variation of well defined macroscopic external fields $a = \{a_j\}(j = 1, \ldots, k)$, and *closed* if energy exchange is not effected via material contact, i.e. particle exchange. (*Open* systems in material contact with the ambience will not be necessary for our consideration).

The basic postulates of statistical mechanics that will be employed by us are : *Postulate P1.* The (classical) probability density distribution f of an isolated equilibrium N_z-body system z is that of the "thickened" microcanonical ensemble (ME), namely [2] (ch. 1.3)

$$f_{z,m}(X_z) = \text{const} \quad , \quad H(X_z, a) \in [E, E + \Delta E];$$

$$f_{z,m}(X_z) = 0 \quad , \quad H(X_z, a) \notin [E, E + \Delta E];$$

$$\Delta E \ll E \quad , \quad \int f_{z,m}(X_z) dX_z = 1$$

[H stands above for the Hamiltonian and $X_z = \{p_z, q_z\}$ –for the set of all momentum (p_z) and position (q_z) variables of system z ; the customary factor $(N_z! h^{3N_z})^{-1}$ in the definition of the phase-space volume element will be inessential for our consideration, so we drop it and define dX_z as $dp_1 \ldots dq_{3N_z}$. If z consists of n sorts of particles ($n > 1$) then, in analogy with $a = \{a_j\}$, N_z should be understood as a complex notation $\{N_{z_i}\}, i = 1, \ldots, n$, etc. We define the constituents of matter so that N_z be constant. This does not rule out processes as e.g. chemical reactions in z since the molecules of z whose number may vary are regarded as composed of the said "elementary" constituents.]

Postulate P2. In a state of thermodynamic equilibrium any N_z-body system $z(N_z = \text{const})$ is assigned an equilibrium statistical entropy given by the information-theoretic formula [3]

$$S_z = -\int f_z(X_z) ln f_z(X_z) dX_z, \tag{1}$$

f_z being the respective ensemble distribution [say, of the ME or the canonical ensemble (CE)].

Postulate P3. The (quasi)static adiabatic processes of phenomenological thermodynamics are described by (quasi)static statistical mechanics.

Some comments on the above postulates and their consequences are necessary at this point.

A well known consequence of P1 is

C1. The density distributions of the equilibrium ensembles (grand canonical, CE, etc) are obtainable via $f_{z,m}$. For instance, if a closed "small" system s under experimental study is in thermal contact with an indefinitely "big" heat bath $b (b \gg s)$, then the (CE) distribution function for s will be $f_{s,c}(X_s) \sim \exp[-H(X_s, a)/\theta]$, where θ is the "reduced" temperature ($= k_B T$) of b and hence of the combined system $s + b$.

The necessity to impart a thermodynamic sense to the statistical entropy $S_{z,m}$ of the ME [eq. (1) with $S_z = S_{z,m}$] requires the validity of the identity

$$dS_{z,m}(E, \Delta E, V, N_z, a) = \frac{1}{\theta_m}(dE + P_m dV - A_m da) \tag{2}$$

($N_z =$ const). The magnitudes in (2) have the usual thermodynamic sense (say, $A_m = \{A_{j,m}\}$ is the set of generalized "forces" corresponding to $a = \{a_j\}$) and subscript m reminds that we have to do with the ME ; analogously, subscript c will refer to CE magnitudes. In particular, the temperature θ_m that eq. (2) assigns to the ME is defined as $\theta_m^{-1} = \partial S_{z,m}/\partial E$.

Another consequence of P1, P2, and C1 is

C2. The equilibrium statistical ensembles (ME, CE, etc) are thermodynamically equivalent.

With respect to ME and CE magnitudes, say, this means that P_m, A_m, etc will differ from P_c, A_c, etc by $o(N_z)$-terms only [for which $o(N_z)/N_z \to 0$ when $N_z \to \infty$]. In particular, extensive magnitudes (proportional to N_z) as entropy coincide up to $O(N_z^{1/2})$-terms [for which $\lim O(N_z^{1/2})/N_z^{1/2} = \text{const} \neq 0$ when $N_z \to \infty$].

Postulate P3 is sometimes explicitly formulated in the literature (cf. e.g. [2], ch. 1.8 and [4], ch. 1.3.4) but some of its consequences do not appear to have been paid the attention they deserve. As these will be important for our consideration, we shall examine them here.

Postulates P1 and P2 and eq. (2) would be of no avail in the absence of P3. Really, their most important product –eq. (2) which combines the first and second law of thermodynamics– must refer to quasistatic *processes* since its phenomenological counterpart envisions exactly such processes. Consequently, statistical mechanics must be capable of establishing a link between the initial and final ME states of z in a given process. The means to achieve this is P3, so we arrive at

C3. There exist quasistatic processes which preserve the ME statistical description of system z for all values of its parameters E, V, N_z, a.

If the process in z is purely adiabatic, then C3 is a direct consequence of P3. If part of dE in eq. (2) is due to heat exchange between z and an external system e, then it is system $e + z$ that should be treated as adiabatic, so P3 will apply to the latter and subsystem z will need special consideration. We shall not have to do, however, with the general case since the combined systems of interest for us are of the kind $b + s(b \gg s)$, at that in a specific context.

It is worth pointing to the fact that classical mechanics possesses a suitable property from the viewpoint of the link between P3 and thermodynamics : the purely dynamic *Liouville theorem* stating constancy with time of phase-space volumes and hence –of the statistical entropy (1) in adiabatic processes has a direct counterpart in the constancy of phenomenological entropy in adiabatic processes.

A simply computable example of the validity of C3 that is of special interest to us is quasistatic adiabatic volume variation of ideal gas : an initial ME description of ideal gas is really preserved at all stages of the process. [The magnitude of ΔE_{id} [cf. eq. (2)] is not constant in this process but what will be important for us is the constancy of $S_{id,m}$ in the process ; the inessential character of ΔE for thermodynamics is noted, say, in [2], ch. 1.6.]

The degree of accuracy of C3 with respect to N_z can be assessed as follows. According to C2, an uncertainty of, say, $O(N_z^{1/2})$ [i.e. $O(N_z^{-1/2})$ *per particle*] for extensive magnitudes makes one uncertain about the concrete (ME, CE, etc) description of z. C3 must therefore be valid for such magnitudes up to terms $O(N_z^{1/2})\epsilon$ per particle, where $\epsilon \ll 1$.

As a preparatory step for the further consideration of P3 we pause here to examine a particular consequences of P1 and P2. By P1 and P2 an isolated system $s + b$, $s \ll b$, possesses a ME distribution $f_{s+b,m}$ and entropy $S_{s+b,m}$. Let s and b by themselves be just closed, i.e.

in thermal contact with each other. Each one of them will then be in equilibrium at the temperature corresponding to the said ME. By P2 each one of them possesses individual entropy (S_s and S_b) of its own defined by eq. (1) for f_s and f_b. Defining $f_{s+b,m}(X_s, X_b)$ as $f_s(X_s)f_b(X_b) + \varphi(X_s, X_b)$ one sees that $S_{s+b,m} = S_s + S_b + S_{cor}$, where the correlation term S_{cor} is due to the presence of the nonfactorable term φ in $f_{s+b,m}$ and cannot be assigned to any one of the systems s and b but pertains to the combined system $s + b$ *as a whole*. The stipulation of well defined individual entropies of s and b therefore requires that S_{cor} should be negligible compared with both S_s and $S_b(S_b \gg S_s$ due to $b \gg s)$. By C1, $S_s = S_{s,c}$ [that is, the CE entropy of s corresponding to the temperature $(\partial S_{s+b,m}/\partial E)^{-1}$ of the ME for $s + b$ (cf. eq. (2))], so the magnitude of S_{cor} cannot surpass $O(N_s^{1/2})$, i.e. $O(N_s^{-1/2})$ per particle. Since $b \gg s$ (i.e. $N_b/N_s \to \infty$, $V_b/V_s \to \infty$), we have $S_{cor}/N_b \sim O(N_s^{1/2})/N_b = [O(N_s^{1/2})/N_b^{1/2}]N_b^{1/2}/N_b = N_b^{-1/2}\epsilon$ ($\epsilon \to 0$ as $N_b \to \infty$). The contribution of S_{cor} per b-particle is thus a higher-order infinitesimal compared with usual "uncertainty" of $O(N_b^{-1/2})$ in equilibrium ensemble description and hence b must be regarded as isolated from s in the limit $N_b/N_s \to \infty$ at $\theta_m = (\partial S_{s+b,m}/\partial E)^{-1} = \text{const}$. Therefore, $f_b(X_b)$ should be that of a suitable ME for b.

The inference just obtained in a purely static statistical picture is an expression of the fact that it would be impossible to arrive at a ME statistical description of the isolated system $s + b$ if $b(b \gg s)$ were not described for all practical purposes by a ME of its own. It is worth demonstrating the vanishing impact of the $s - b$ correlations on the ME description of b in the above limit by means of a pertinent nonstatic statistical process as well that does not violate the static character of the phenomenological picture. Assume to this end that s and b are initially isolated from each other and from the rest of the Universe. By P1, each one of them is described then by a ME of its own. Assume too that $\theta_{s,m} = (\partial S_{s,m}/\partial E_s)^{-1} = \theta_{b,m} = (\partial S_{b,m}/\partial E_b)^{-1}$ and "switch on" at moment $t = 0$ thermal contact between s and b, keeping s and b *closed* and isolated from the rest of the Universe. From the viewpoint of statistical mechanics we shall have at $t > 0$ a "thermodynamically hidden" process (at a temperature $\theta = \text{const} = \theta_{s,m} = \theta_{b,m}$) in which, by P3 and C1, the initial ME for s will be eventually transformed into a CE for s at the same θ, so $S_s(t \to \infty) = S_{s,c} = S_{s,m} + O(N_s^{1/2})$. Due to the Liouville theorem we have $S_{s+b}(t > 0) = \text{const}$, which immediately leads to the above inference that, up to terms generating only higher-order infinitesimals $\sim N_b^{-1/2}\epsilon$ in extensive b-magnitudes, the statistical description of b at $t \to \infty$ is given by a pertinent ME.

The said inference is easily extended to the case of quasistatic adiabatic *variation* of (at least some of) the mechanical-type parameters of $s+b$ [denoted by V and a in eq. (2)] when s and b are in thermal contact with each other. Indeed, by P3 s and b are in thermal equilibrium at each moment t (so that s possesses a CE statistical description at each t) and by C3 the combined system $s+b$ possesses –up to higher-order infinitesimals– an ME description at any moment t when the initial condition at $t=0$ is also given by an ME for the said system. We therefore have essentially the same situation here as the one discussed in the purely static case, so one arrives at

C4. Up to higher-order infinitesimals the heat bath $b(b \gg s)$ possesses an ME statistical description at any moment $t>0$ when system $s+b$ undergoes a quasistatic adiabatic process at positive times under an ME initial condition at $t=0$.

As a last preparatory step we shall now discus the

3. Interpretation of ideal-gas entropy variation as variation of randomness

As is well known, the right-hand side of eq. (2) is equal to dQ_m/θ_m, dQ_m standing for the heat energy imparted to the system in the process. This equation has its CE counterpart which reads, for an only variable parameter V (cf.[1]),

$$dS_c = \theta_c^{-1}(d\langle H\rangle_c + P_c dV) \equiv \theta_c^{-1} dQ_c \qquad (3)$$

in obvious notations, $d\langle H\rangle_c$ denoting, in particular, the CE variation of the average (internal) energy $U = \langle H\rangle_c$ of the system. In the case of an ideal-gas N-body system the energy E_{id} of its dynamic state is equal to its kinetic energy K and one obtains

$$d\langle H_{id}\rangle_c = (\partial\langle K\rangle_c/\partial\theta_c)d\theta_c$$

(since $\langle K\rangle_c$ is V-independent at a fixed θ_c). The respective entropy variation $dS_{id,c}$ will therefore be give by

$$dS_{id,c} = \theta_c^{-1}(\frac{d\langle K\rangle_c}{d\theta_c}d\theta_c + P_c dV) \qquad (4)$$

Other CE ideal-gas equations that will be employed below are

$$P_c V = N\theta_c \tag{5}$$

$$\langle K\rangle_c = (3/2)N\theta_c = N\langle\vec{p}^2\rangle_c/2m \tag{6}$$

Consider first an infinitesimal variation dV of the ideal-gas volume V at $\theta_c = \text{const}$. Eqs. (4) and (5) yield in this case entropy variation

$$d_V S_{id,c} = (P_c/\theta_c)dV = NdV/V \tag{7}$$

This expression agrees well with an intuitively acceptable concept of position-randomness variation in an N-body system with statistically independent positions when one possesses, at that, no information whatsoever about the location of any given individual particle (as is the case with the ideal-gas CE position distribution). Really, the magnitude of volume V represents a natural measure of the indefiniteness of position location of any individual particle. Ratio dV/V on its turn measures –with the proper sign– the relative significance of the said variation and can be treated as an "absolute" measure of individual position-randomness variation. The magnitude NdV/V in eq. (7) can therefore be treated as position-randomness variation of the *gas* in accord with the natural concept that the position-randomness variation in a system of N identical particles with statistically independent positions should represente the sum-total of equal individual contributions due to the identical statistical properties of each individual particle. Consequently, in this sense $d_V S_{id,c}$ represents a measure of position-randomness variation dR_{pos} in the ideal gas.

Examine now the variation $d_{\theta_c} S_{id,c}$ at $V = \text{const}$. Eq. (4) yields in this case $d_{\theta_c} S_{id,c} = \theta_c^{-1}(d\langle K\rangle_c/d\theta_c)d\theta_c$, whence one obtains with the aid of (6)

$$d_{\theta_c} S_{id,c} = Nd\langle\vec{p}^2\rangle_c^{3/2}/\langle\vec{p}^2\rangle_c^{3/2} \tag{8}$$

The form of eq. (8) is the same as that of eq. (7) thus implying that the right-hand side of (8) represents momentum-randomness variation dR_{mom} in the system of N statistically independent individual momenta $p_i, i = 1, 2, \ldots, N$, of the ideal gas. The randomness interpretation of $dR_{mom} = d_{\theta_c} S_{id,c}$ is additionally corroborated by the fact that dR_{mom} and dR_{pos} are magnitudes of an *identical physical nature* in the sense that each one of them can be transformed in a strictly dynamic (mechanical) fashion into an equivalent amount of the other and, at the same time, the meaning of randomness is already fixed by our interpretation of the magnitude NdV/V.

(The above definition of equivalence is in keeping with a natural physical concept concerning mutually transformable magnitudes, e.g. with the equivalent physical nature –energy– of kinetic and potential energy variations in a conservative dynamic system.)

In order to demonstrate the mutual dynamic transformability of dR_{pos} and dR_{mom} into each other one has to examine a mechanical process in the ideal gas in which entropy variation $dS_{id} = dR_{mom}+dR_{pos}$ is zero, i.e. $dR_{mom} = -dR_{pos}$. This will evidently be a quasistatic adiabatic process. The CE is not quite suitable to this end due to the contact of our system with the thermostat in the said picture, so we shall consider the problem in the ME picture and then make use of C2 for these ensembles.

For ideal gas with energy $E_{id} = K$ and only variable parameter V eq. (2) reads

$$dS_{id,m} = (1/\theta_m)(dK + P_m dV) \tag{9}$$

Comparing eqs. (9) and (4) one sees that if the kinetic energy K in $S_{id,m}(K,V)$ is set equal to $\langle K\rangle_c$, the thermodynamic equivalence of the ME and the CE (C2) will lead to

$$\theta_m = \theta_c = \theta \quad , \quad P_m = P_c = P \tag{10}$$

in the thermodynamic limit.

Eqs. (10) evidently mean that $d_V S_{id,m}$ and $d_\theta S_{id,m}(= dK/\theta)$ have the same meaning and value (up to "infinitesimals") in the thermodynamic limit as $d_V S_{id,c}$ and $d_\theta S_{id,c} = d\langle K\rangle_c S_{id,c}$, i.e. variations of dR_{pos} and dR_{mom} respectively. On the other hand, we know (both from C3 and direct computation) that the ME statistical description of ideal gas is preserved in a process of quasistatic adiabatic variation of its volume V. This fact, together with the "entropy conservation law" $dS_{id,m} = dR_{pos} + dR_{mom} = 0$ in the said process, has at that a purely mechanical sense being a consequence of microscopic dynamics when applied to an ME of ideal-gas mechanical states. Consequently, the identity $dR_{pos} = -dR_{mom}$ in such processes represents a dynamic transformation law. As is clear e.g. from eq. (9) (with $dS_{id,m} = 0$), the said law directly follows from the enertgy conservation law of the overall system ideal gas + rest of the Universe. Consequently, dR_{pos} and dR_{mom} are magnitudes of the same physical nature (randomness –the clear-cut nature of dR_{pos}). The same applies therefore to the sums in eqs. (4) and (9), which are generally $\neq 0$.

The consideration in this section gives a precise definition of the meaning of *randomness variation* in our particular approach : this is nothing else than ideal-gas nonzero statistical entropy variation in the CE and/or ME pictures. If this entropy variation could be imparted to an arbitrary system s by an ideal-gas heat bath in a purely dynamic, conservative and ensemble-statistics preserving fashion, then, employing the third law assigning zero entropy to s at $\theta = 0$, one could achieve a randomness interpretation of any S_s and its variation. As we shall see now, our axioms permit entropy exchange of this kind.

4. The interpretation

We shall examine here a combined system $s + b_{id}$ consisting of an arbitrary system s under experimental study and ideal-gas heat bath $b_{id}(b_{id} \gg s)$. b_{id} and s are assumed to be closed and in thermal contact with each other, the combined system $s+b_{id}$ being adiabatically isolated from the rest of the Universe.

(i) Let the initial condition for $b_{id} + s$ at $t = 0$ be thermodynamic equilibrium described by a pertinent classical ME. Assume that the parameters V_s and a_s of s and V_{id} of b_{id} are varied quasistatically and adiabatically at $t > 0$. (The variation of V_{id} will ensure temperature variation in $s+b_{id}$, if necessary.) By C3, the ME description of $s+b_{id}$ is preserved in the process and by C4 the same applies (up to higher-order infinitesimals) to b_{id}. By the Liouville theorem, $S_{b_{id}+s}$ is constant in the process, so we have a dynamic entropy conservation law in $b_{id} + s$ of the kind discussed in Sec. 3. Up to higher-order infinitesimals the entropy exchange (of magnitude proportional to N_s) between s and b_{id} can be represented as $\Delta S_{s,c} = -\Delta S_{b_{id},m}$. Since $\Delta S_{b_{id},m}$ has a randomness interpretation, the same interpretation applies to $\Delta S_{s,c}$ too (cf. Sec. 3). By C2, the same interpretation applies to any equilibrium ensemble description of s. This result is general and is also valid when we have, say, chemical reactions and phase transitions in s.

(ii) Assume now that the classical character of b_{id} is preserved but the processes in s require quantal description. We impose then quantal description of $b_{id}+s$. Examine once again the above adiabatic process in $b_{id}+s$. The well known quantum variant of the Liouville theorem states that $\Delta S_{b_{id}+s} = 0$ in the process, so the dynamic ”entropy conservation law” operates in this case too. The quantal version of postulates P1-P3 requires, in complete analogy with the classical case, an ME description of $b_{id} + s$ at all times $t > 0$ and hence –up to higher-order infinitesimals– an ME description of b_{id}. The classical character of b_{id}, however, admits a classical ME description of b_{id} as well and hence a randomness interpretation of the entropy variation in s.

5. Conclusions

Our consideration resting on postulates P1-P3 and the Liouville theorem demonstrated that a randomness interpretation of entropy of a most general character is possible indeed. (Invalidity of some of these postulates would mean that not only our interpretation but statistical mechanics itself too rest on a shaky basis.) With respect to this interpretation the ideal-gas heat bath really turned out to play a role as significant as that of the ideal-gas thermometer in temperature measurements. The above way of reasoning gives also an answer to the question why purely mathematical information-theoretic expressions for statistical entropy of the kind of (1) have a thermodynamic purport : these entropies coincide up to $O(N_z^{1/2})$ and are transformable in a conservative fashion into ideal-gas ME entropy, the latter having a lucid randomness sense, at that directly linked with ideal-gas thermodynamics.

Our interpretation would also be preserved in a possible more general future theory containing quantum and classical statistical mechanics as its particular limits if this theory would contain, in its specific language, postulates P1-P3 and a Liouville-type theorem stating constancy of entropy in adiabatic processes.

An obvious by-product of our consideration that deserves mention too is that for compound systems of the kind $b + s$, $b \gg s$, we have an ME statistical variant (2) of the basic thermodynamic identity for b and a CE variant $dS_{s,c} = \theta_c^{-1}(dU_c + P_c dV - A_c da)$ of the same identity for s. (In fact, it is exactly the latter variant that is made use of in ref. [4].)

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