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Temperature, Entropy, and Kinetic Theory

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The relation between the concepts of temperature and entropy and the kinetic theory of gases is discussed, with particular attention to the aspects which are frequently treated as obvious or not even mentioned. In order to show that the usual thermodynamic relations are by no means obvious and may be contradictory, the model of a discrete velocity gas is used. It is also shown that the usual relation between the entropy rate and the heat supplied to a gas is not valid (even close to equilibrium) unless the theory is Galilei invariant (which is obviously not the case for a discrete velocity gas) and must be replaced by another one that eliminates all the paradoxical aspects of the matter.

KEY WORDS: Temperature; entropy; kinetic theory.

1. INTRODUCTION

The fact that this paper is meant to contribute to the celebration of Matthieu Ernst's 60th birthday gave me a hint to the topic to choose. I am, in fact, sure of his interest in this topic. As will be indicated later, we even corresponded once on a particular aspect of the present paper.

What new can be said about the subject indicated in the title? We start by recalling a few well-known facts, in order to fix the notation.

The second law of thermodynamics states, as is well known, that there is a state function S, called entropy, such that (in the case of a fixed amount of gas, to which we shall stick here):

(a) In any reversible transformation

$$T\dot{S} = \dot{E} + p\dot{V} \tag{1.1}$$

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where the dot denotes a time derivative, E the energy of the system, T the absolute temperature, p the pressure, and V the volume occupied by the gas.

(b) In any irreversible transformation

$$T\dot{S} \geqslant \dot{E} + p\dot{V} \tag{1.2}$$

Perhaps we should add a word of explanation here. If one opens any book on thermodynamics, one will find the above relations written in a slightly different way, i.e., with the time derivatives $\dot{S}$, $\dot{E}$, $p\dot{V}$ replaced by the differentials dS, de, dv. The two formulations are, of course, completely equivalent, because the first differential of a dependent variable is nothing else than the first derivative multiplied by the increment (differential) of the independent variable. The latter, in dynamics, is usually time; however, time is rarely, if ever, mentioned in thermodynamics, to the point that somebody can dub the latter as "thermostatics." The notation with differentials is very convenient when one wants to study thermodynamics as the science dealing with equilibrium states and state functions. A very elegant discipline arises, which is more mathematical than physical and can be likened to Euclidean geometry for its beauty and self-consistency. In physics, however, the states dealt with in this discipline are reached through time evolution and so there is a thermodynamics of processes, which requires time derivatives. These processes are usually time irreversible and thus a special discipline, called thermodynamics of irreversible processes or thermomechanics, has grown to describe them. In this case the use of time derivatives is imperative. As a matter of fact, even usual books of thermodynamics, if they write Eq. (1.2) at all in terms of differentials, risk confusing the reader, unless they make it clear that time is the independent variable. Why should entropy increase in an isolated system if we choose another variable, say volume or pressure or, indeed, entropy itself, as an independent variable?

Here we study the connection between kinetic theory and thermodynamics and thus the use of time derivatives is also essential. Before entering this topic, however, we should mention the relation between the two kinds of thermodynamics that we have just mentioned. The thermodynamics expounded in the books that bear its name in the title (the one dealing with state functions) can be recovered from the thermodynamics of processes in the limit of very slow processes when inhomomogeneities, higher order time derivatives, and squares of time derivatives can be neglected.

Although temperature is an intuitive concept, known from everyday life, and entropy a rather difficult concept, we face the following paradoxical situation: entropy (at least for a monatomic perfect gas) is well defined in kinetic theory (as the integral of $f \log f$) for both equilibrium and nonequilibrium states, temperature is not. Temperature is only defined for a system in equilibrium; this does not prevent us, of course, from extending the concept to nonequilibrium states. This can be done by either assuming a local equilibrium or, using some basic relation holding in equilibrium, to *define* temperature for nonequilibrium states as well. In fact, temperature in kinetic theory usually appears in connection with internal energy per unit mass e or pressure p. Here are two typical relations for a monatomic perfect gas:

$$e = \tfrac{3}{2}RT; \qquad p = \rho RT \qquad (R = k/m) \tag{1.3}$$

Here k is the Boltzmann constant and m the mass of a molecule. This is justified by known laws holding in kinetic theory for monatomic perfect gases. We remark that e, ρ, and p can be defined out of equilibrium as well through moments of the distribution function.

However in thermodynamics there is no reason why both relations should hold at the same time. The only thing that one can show is that if the second relation in (1.3) is assumed to hold, then e must be a function of T only, but there is no reason why this relation should be linear. Sometimes one talks of a thermally perfect gas when the second equation in (1.3) holds and of a calorically perfect gas when the first one does.

If we treat deviations from equilibrium (velocity, heat flow, deviation of the stress tensor from isotropic pressure) as of order ε (a smallness parameter) and neglect terms of order ε^2, then we can easily obtain from the Boltzmann equation the first principle of thermodynamics, in the form

$$\dot{E} = -p\dot{V} + q \tag{1.4}$$

where E is the total energy, V the volume, and q the heat flowing into the gas per unit time. This is obtained from the energy balance equation with due care in evaluating the derivatives when the domain changes in time.

The concept of temperature is made unique, except for a constant factor, by the second principle. Usually the identification of T is made trivial in kinetic theory, sometimes reduced to a definition.

But this definition might be wrong, in the sense that it might contradict some accepted ideas or relations. To see this we shall consider the example of the lattice gas (discrete positions, velocities, and time) or, in order to avoid problems related to volume changes, a merely discrete velocity gas (with continuous space-time).

As a matter of fact, my interest in the topic of the present paper was spurred by a talk by Ernst,$^{(1)}$ who a few years ago pointed out that "many authors have introduced quantities, called "temperatures" of a Cellular

Automata fluid, that do not coincide with the true temperature in the sense of thermodynamics and statistical mechanics." Actually, his criticism specifically applies to the kinetic theory of discrete-velocity gases, because it is not the discretization of space variables, but the discrete nature of velocities that plays a significant role in the circumstance that certain thermodynamic relations change when we leave the confortable realm provided by the traditional gas with a continuous set of velocities.(2, 3) As a matter of fact, in the same volume of Proceedings where Ernst's paper(1) appeared there is another paper,(4) where a definition of temperature at variance with that supplied by statistical mechanics is used. The authors can be perhaps excused because this debatable definition has become traditional since the pioneering treatment of Gatignol;(5) even a more recently published book,(6) however, contains the same arbitrary choice, without justifying it.

As Ernst pointed out, the gas temperature T is something that appears in the equilibrium canonical distribution, proportional to $\exp(-\beta E)$, where E is the total energy of the gas; precisely, as equilibrium statistical mechanics teaches us, $\beta = 1/(kT)$. It is amusing to see that the authors of the above-mentioned paper(4) devote some effort to the problem of relating β and T (defined in a different way); their relation is strongly nonlinear and complicated.

The aim of this paper is thus to discuss the thermodynamics of a gas in terms of kinetic theory with particular reference to the concepts of temperature and entropy. The case of a discrete velocity gas will show why this is necessary. In fact, at first, it would seem that there is no need for such an enterprise because everything is solved by the simple remark by Ernst. It seems appropriate, however, to discuss this matter from the viewpoint of the kinetic theory of gases, without any reference to general results of equilibrium statistical mechanics. This is also convenient from the point of view of a gas with a continuous distribution of velocities, because it shows what subtleties are hidden under certain statements that are frequently introduced in the form of mathematical definitions.

We shall find, in fact, that some familiar thermodynamic relations cannot be applied at all, with the consequence that one should be careful before talking about the "thermodynamics of a discrete-velocity gas."

2. BOLTZMANN EQUATION, STATE VARIABLES, AND EQUATIONS OF STATE.

According to standard definitions, a discrete velocity model of a gas is a system of partial differential equations of hyperbolic type (discrete Boltzmann equation), having the following form:

$$\partial f_i/\partial t + v_i \cdot \partial f_i/\partial x = Q_i(f, f) \tag{2.1a}$$

$$Q_i(f, f) = \sum_{j,k} c_{ijk} k_j f_k - \sum_j k_{ij} f_j f_i \tag{2.1b}$$

where v_i are the discrete velocities (vectors of R^3) belonging to a prearranged discrete set, c_{ijk} and k_{ij} are positive constants, and the indices run from 1 to p, while f_i ($i = 1, 2,\ldots, p$) are the probabilities (per unit volume) of finding a molecule at time t at position x with velocity v_i. We shall write f, as done in (2.1b), for the collection f_i.

We certainly must assume that the collision term $Q_i(f, f)$ satisfies the restrictions needed to guarantee the conservation of mass, momentum, and energy and the entropy inequality. We remark that this would be a generalization with respect to the traditional concept of a discrete velocity gas,$^{(5)}$ where it is assumed that each single collision satisfies momentum and energy conservation. This restriction, dictated by a strict analogy with the case of continuously distributed velocities, is rather inconvenient. In fact: (a) the traditional models are very restricted in number and frequently plagued with accidental degeneracies, such as the presence of more than five collision invariants and hence additional conservation laws with respect to the traditional ones (mass, momentum, and energy); (b) their extension to mixtures seems hard if not impossible, especially when the ratio of masses is irrational (in fact I do not know of any discrete-velocity model of the traditional kind for mixtures, with the exception of the case when all the momenta have the same magnitude and hence conservation of energy follows from the conservation of the number of particles); (c) there is not a known procedure to approximate in arbitrary fashion the continuous case by a sequence of discrete models, at least if we want to satisfy all the formal properties of the Boltzmann equation, because (in more than two dimensions) the known models satisfying the conservation equations and the H-theorem and not degenerate for some reason are finite in number.

However if we only assume that

$$\sum_i Q_i(f, f)\, \psi_{\alpha i} = 0 \qquad (\alpha = 0, 1, 2, 3, 4, 5) \tag{2.2}$$

where ψ_α ($\alpha = 0, 1, 2, 3, 4, 5$) are the five collision invariants $(1, \xi_1, \xi_2, \xi_3, |v|^2)$, where $\xi_j = (j = 1, 2, 3)$ are the Cartesian components of v, we lose several important properties of the Boltzmann equation. The physical interpretation of these extended discrete-velocity models would be the following. A collision is a more complicated process than in the continuous velocity model. When two particles meet they undergo a not completely deterministic process, in the sense that we cannot guarantee that another pair will emerge from the collision with certain velocities but only that the precollision momentum and energy will be distributed with a certain probability to a number of pairs; this is true even in the continuous Boltzmann equation because the collision parameters also determine the postcollisional velocities. Here we go a step further because we do not exhibit possible pairs of these velocities; thus there are no elementary processes where mass, momentum, and energy are conserved, but we only ensure that momentum and energy are conserved globally.

Unfortunately, if we also assume the validity of an H-theorem, this can be shown to require the conservation of momentum and energy in each single collision. As a consequence, however, we can also show the existence of a "Maxwellian" distribution $\mathcal{M}_i$ such that $Q(\mathcal{M}, \mathcal{M})=0$ and $\log \mathcal{M}$ is a linear combination of the collision invariants. Thus we must stick to traditional discrete models.

The macroscopic quantities are computed, as usual, by taking averages, weighted with f_i, of the microscopic quantities. Thus the density ρ is obtained from

$$\rho(x, t)=\sum_i f_i(x, v_i, t) \tag{2.3}$$

A discussion of the momentum balance leads to the result that the pressure of the gas should be computed through

$$p(x, t)=(1/3)\sum_i c_i^2 f_i(x, v_i, t) \qquad (c_i=v_i-u) \tag{2.4}$$

where u is the bulk velocity and the factor 3 arises from the fact that space is three-dimensional and we assume that an interchange of the coordinates leaves the gas equations invariant. Balance of energy indicates that the energy per unit mass e should be defined by means of the following relation:

$$\rho e(x, t)=(1/2)\sum_i c_i^2 f_i(x, v_i, t) \tag{2.5}$$

All these definitions are exactly the same as in the case of a gas with continuous velocities, the only difference being that sums over a discrete set replace integrals. Actually what we shall say applies to continuous velocities as well. In this case, as we shall see, both relations in (1.3) will turn out to hold by what appears to be a coincidence, but, as we shall see, is subtly related to the Galilei invariance of the Boltzmann equation, which, of course, is not available in the case of discrete velocities.

It is an obvious consequence of Eqs. (3)–(5) that the following equation of state holds:

$$p = (2/3)\, \rho e \tag{2.6}$$

This equation relating density, pressure and internal energy holds for any dilute "monatomic" gas, i.e. any gas made of molecules, whose internal and potential energies are negligible. At this point, it is tempting to introduce temperature(4–6) by means of

$$e = (3/2)\, RT \tag{2.7}$$

in order to obtain the usual equation of state $p = \rho RT$. This step is purely formal and is not related at all to the physical meaning of temperature.

3. THE USUAL RELATIONS MAY PRODUCE A CONTRADICTION.

In order to introduce the concept of temperature, we remark that this name is used for a quantity having the property of having the same value for two systems of particles which are able to exchange energy and are in an equilibrium state. This definition implies that we must define temperature for equilibrium states; the extension of this concept to nonequilibrium gases can be performed only by assuming that the same relations holding among internal energy, pressure, density and temperature in an equilibrium state, remain true out of equilibrium as well. The same definition also shows that it would be inappropriate to try to define temperature without introducing at least two subsystems of particles, the molecules of the first subsystem being distinguishable from those of the second one for at least one (observable) physical property.

The simplest property which can be used to mark the two subsystems is the mass possessed by each molecule of the subsytem. We thus might think of considering a mixture of two gases in a state of equilibrium. Unfortunately, this is easy for continuous-velocity gases, but poses problems for discrete models because of the remark (b) in the discussion following Eq. (2.1b) in Section 2.

We thus (somehow artificially) accept, in order to avoid a lengthy discussion which would lead to the same result, that a discrete velocity in equilibrium at rest with a uniform temperature T has a Maxwellian distribution (here, as usual in the literature on discrete-velocity gases, and, as we already did, using quotes, in the previous section, we call Maxwellian the equilibrium distribution which makes the collision term vanish and must be, because we assumed that the H-theorem also holds, an exponential of a linear combination of collision invariants). We denote the Maxwellian for a gas at rest by

$$\mathcal{M}_i = A \exp(-\beta m v_i^2/2) \tag{3.1}$$

where m is the molecular mass of the gas and β must be a universal function of the temperature T; the precise relation between β and the temperature cannot be obtained, however, without introducing the second law of thermodynamics. The latter is expressed by Eqs. (1.1) and (1.2). In other words the second law defines T uniquely (except for a constant factor), whereas, if we adopt the point of view that temperature is just an index of equilibrium between two subsystems and T is such a quantity, then any function of T, $\Theta(T)$, is another such quantity.

The questions are: What does kinetic theory exactly tell us about the second law? Can we apply the relations holding in thermodynamics in kinetic theory (in particular, to a discrete-velocity gas)? If not, is there a thermodynamics for such a system?

In order to answer these questions, we try to assume that the relations (1.1) and (1.2) can be applied and see whether we arrive at sensible results. To this end, we must exploit the well-known circumstance that the quantity corresponding to entropy in kinetic theory is $-RH$, where H is the Boltzmann functional; here R is a constant, to be later identified with a universal constant k (Boltzmann's constant) divided by the molecular mass. Then if we let

$$H = \int \sum_i \log \mathcal{M}_i \mathcal{M}_i \, dx \tag{3.2}$$

a candidate to entropy is $S = -H/R$. Thus in a (homogeneous) equilibrium state we should have

$$S = -R \int \sum_i \log \mathcal{M}_i \mathcal{M}_i \, dx \tag{3.3}$$

$$M = \int \sum_i \mathcal{M}_i \, dx \tag{3.4}$$

$$E = (1/2) \int \sum_i c_i^2 \mathcal{M}_i \, dx \tag{3.5}$$

where M is the total mass and $\mathscr{M}_i$ a Maxwellian distribution given by

$$\mathscr{M}_i = A \exp(-\beta m c_i^2/2) \tag{3.6}$$

In addition, $c_i = v_i$ if we assume the gas to be at rest. Thus

$$S = R\beta m E - RM \log A \tag{3.7}$$

$$M = AV \sum_i \exp(-\beta m c_i^2/2) \tag{3.8}$$

$$E = AV(1/2) \sum_i c_i^2 \exp(-\beta m c_i^2/2) \tag{3.9}$$

In a change which is reversible and hence goes through equilibrium states we have, by differentiating Eqs. (3.7) and (3.8) (since the total mass M remains constant),

$$\dot{S} = R\dot{\beta} m E + R\beta m \dot{E} - RM\dot{A}/A \tag{3.10}$$

$$0 = M\dot{A}/A + M\dot{V}/V - m\dot{\beta}E \tag{3.11}$$

and, as a consequence, by eliminating $\dot{A}$,

$$\dot{S} = R\beta m \dot{E} + RM\dot{V}/V \tag{3.12}$$

By comparing this with Eq. (1.1), we find

$$\beta = 1/(kT), \qquad p = RTM/V = R\rho t \tag{3.13}$$

where $k = Rm$ must be a universal constant (because of our assumption that β is a function of just temperature). This gives not only the desired identification of the temperatures but also the state equation for a perfect gas. The latter result, is however contradictory because, when put together with Eq. (1.3), which always holds, it gives Eq. (2.7). This is of course satisfactory for the usual Boltzmann gas with continuous velocities because

$$3/(2\beta m) = (1/2) \int c^2 \exp(-\beta m c^2/2)\, dc \bigg/ \int \exp(-\beta m c_i^2/2)\, dc \tag{3.14}$$

is an identity, but in the case of a gas with discrete velocities it would imply

$$3/(2\beta m) = E/M = (1/2) \sum_i c_i^2 \exp(-\beta m c_i^2/2) \bigg/ \sum_i \exp(-\beta m c_i^2/2) \tag{3.15}$$

i.e., for any value of the parameter b

$$3/(2b) = \sum_i c_i^2 \exp(-bc_i^2) \Big/ \sum_i \exp(-bc_i^2) \tag{3.16}$$

which is obviously absurd, unless we go to the limit of infinitely many velocities with a continuous distribution.

One may try to escape from this difficulty by modifying the definition of S slightly, e.g., by taking, as permitted, $\mathscr{S} = S - R\beta Me$ in place of S, but the conclusion is similar.

4. THE UNUSUAL THERMODYNAMICS OF A DISCRETE-VELOCITY GAS.

I had a chance of mentioning the difficulties discussed in the previous section to M. Ernst via e-mail, and he pointed out to me that he had been worrying about this problem since 1989 when he was lecturing in a summer school. His lectures notes have since been published.(7) His suggestion is that one should not hasten to identify the kinetic and thermodynamic pressures.

But how should one identify thermodynamic pressure in kinetic theory? Thermodynamics has to do with conservation of energy (first law) and irreversibility (second law). Both aspects are present in kinetic theory. If we consider a gas in a rectangular box with a movable wall in a state with isotropic stresses (given by the kinetic pressure), the balance equation for energy is given by Eq. (1.4).

Concerning q, one has the basic result first indicated by Darrozès and Guiraud (see ref. 8), which is of paramount importance when discussing the H-theorem in the presence of walls and can be stated in the following form:

$$\int v \cdot n f \log f \, dv \leqslant -\beta_w \int v \cdot n \, |v|^2 f \, dv \qquad (x \in \partial\Omega) \tag{4.1}$$

Equality holds if and only if f coincides with $\mathscr{M}_w$ (the Maxwellian with the temperature of the wall) on the boundary $\partial\Omega$ (unless the scattering kernel in the boundary conditions is a delta function).

We remark that if the gas does not slip upon the wall, the right-hand side of Eq. (4.1) equals $-q_n/(RT_w)$, where q_n is the heat flow along the normal. If the gas slips on the wall with velocity u, then one must add the power of the stresses $p_n \cdot u(=p_{ij}n_i u_j)$ to q_n. In this case, however, the right-hand side of Eq. (4.1) still equals $-q_n^{(w)}/(RT_w)$, where $q_n^{(w)}$ is the heat flow in the solid at the wall. In fact $q_n + p_n \cdot u = q_n^{(w)}$ because the normal energy flow must be continuous through the wall (unless there are energy sources concentrated at the surface) and stresses have a vanishing power in the solid, since the latter is assumed to be at rest.

This result, holding in kinetic theory with continuous velocities(2) is trivially extended to the case of discrete velocities. Then the time rate of $H = -RS$ is related to q through

$$\dot{H} \leqslant -q/RT_w \tag{4.2}$$

where T_w is the temperature of the boundary through which the heat is supplied, provided that the boundary is at rest. The inequality sign turns into an equality when the irreversible processes associated with molecular collisions may be neglected; in other words when the deviation of the distribution function from a Maxwellian is negligible. Equations (4.2) and (1.4) give

$$T_w \dot{S} \geqslant \dot{E} + p\dot{V} \tag{4.3}$$

This is to be compared with Eq. (1.2). In fact, if we consider a very slow process such that the gas temperature T equals T_w and the irreversible processes in the gas may be neglected (in other words, if the deviation from a Maxwellian distribution with temperature T_w is negligible), then Eq. (3) coincides with Eq. (1.2). Thus there seems to be no way out of the difficulties. However, as remarked by the author in a previous paper,(8) there is a *proviso* in the above, i.e., that the boundary does not move. This is clearly incompatible with the fact that in the above relations we must allow the volume to change. Let us then reexamine the result (4.2) in the light of this remark. In the case of a moving boundary, Eq. (4.2) is replaced by

$$\dot{H} \leqslant -\left[q - \int (w_n - u_n)\, p\, d\sigma \right] \Big/ RT_w + O(v^2) \tag{4.4}$$

where w is the vector appearing in the expression of a Maxwellian

$$\mathscr{M}_i = A \exp[-(v_i - w)^2/2RT_w] \tag{4.5}$$

and the integral is extended to the moving part of the boundary; the subscript n denotes the normal component (with the normal oriented from the boundary into the gas). In the case of continuous velocities, one easily shows (by a direct calculation or by Galilean invariance) that $w = u$. This is not the case for a discrete-velocity gas. In fact now we easily find

$$u = pw/(\rho RT) + O(w^2) \tag{4.6}$$

The terms of order $u^2 (= O(w^2))$ can be neglected because we consider an extremely slow process. Then Eq. (4.4) becomes

$$\dot{H} \leqslant -\left[q - \int (\rho RT - p)\, u_n\, d\sigma \right] \Big/ RT_w \tag{4.7}$$

or, since ρ, T, p are assumed to be uniform on the boundary,

$$\dot{H} \leqslant -[q + (\rho RT - p)\, \dot{V}]/RT_w \tag{4.8}$$

since the integral of u_n over the interface is $-\dot{V}$. Hence assuming a reversible process [and ignoring the (by now) inessential subscript in T_w]

$$T\dot{S} = q + (\rho RT - p)\, \dot{V} = \dot{E} + \rho RT \dot{V} \tag{4.9}$$

In other words the second law is satisfied with the definition of temperature arising in statistical mechanics, but the change of entropy in a reversible process is not simply related to the heat supplied to the gas! It also depends on the rate of change of the volume. Of course this term disappears when we have a Galilei invariant theory, which is the case for a continuous velocity gas.

5. CONCLUDING REMARKS

We have discussed the relation between the concepts of temperature and entropy and the kinetic theory of gases and we have shown that the usual thermodynamic relations are by no means obvious and may be contradictory in some cases. The model of a discrete-velocity gas has been used as an example to illustrate the possible pitfalls. It has also been shown that the usual relation between the entropy rate and the heat supplied to a gas is not valid (even close to equilibrium) unless the theory is Galilei invariant (which is obviously not the case for a discrete-velocity gas) and must be replaced by another one that eliminates all the paradoxical aspects of the matter. We have also discussed the difficulties of discrete-velocity gases, such as the lack of a general extension to mixtures, the impossibility of finding a sequence of them that arbitrarily approximates a gas with continuously distributed velocities, and the possible lack of some collision invariants or (on the contrary) the presence of spurious collision invariants.

Of course, at this point one might wish to know more. What is the situation with a dense gas, a liquid, or a solid? The problem here is that we do not have a kinetic theory as accurate as we have for a rarefied gas. Even in the case of hard spheres, the Enskog equation is rather accurate, but not exact, because it omits memory effects. Thus we do not have the tools to discuss the problem as accurately as we have done for a rarefied gas.

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