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THERMODYNAMICS OF NEGATIVE ABSOLUTE PRESSURES

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States with negative absolute pressure are investigated from thermodynamic viewpoint. It is found that negativity of pressure does not contradict Callen's postulates, and the postulates cannot be extended in a natural way to rule out just these states. These states may be stable against small fluctuations. In nuclear physics, QCD and GUT, $p < 0$ states are easy to interpret by thermodynamics.

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1. Introduction

While pressures of all *familiar* states of matter are non-negative, negative pressures have recently become important in some exotic but fundamental physical situations. Here we only list three such ones, as: (*i*) dilute cool nuclear matter, (*ii*) dilute cool quark-gluon plasma, (*iii*) supercooled symmetric phase of Grand Unification. (Some more details, of course, will be given in Sect. 2.) Calculations indicate that matter can indeed enter the negative pressure region in Case (*i*), and the present state of Universe seems to be hardly understandable without a prehistory in state (*iii*).

On the other hand, there is some reluctance and uneasiness in the literature when meeting such states. There is a whole spectrum of opinions, and they are seldom completely cathegoric, still, it is often said that states of negative pressure are not fully realistic. As a moderate example, consider a statistical physics textbook with extensive thermodynamical discussions [1]. It has three comments on $P < 0$ states. In § 12 of Ref. [1] it is said (translated to English by one of us): "It is easy to see that the pressure of a body is necessarily positive in any equilibrium state ... Nevertheless, there is an essential difference between Conditions $T > 0$ and $P > 0$. Bodies of negative temperature would be completely unstable and cannot exist in Nature. On the other hand, nonequilibrium states of negative pressure may exist in Nature with limited stability". The book calls $P < 0$ states *metastable* compared to both familiar *stable* states and $T < 0$ *unstable* ones. Finally, later, it is told that overheated liquids may have metastable $P < 0$ states.

Now, there can be no doubt that $P < 0$ states do indeed have a tendency for droplet formation [1, 2, 3]. However, this instability of the *flow* may appear on time scales quite long compared to those of thermodynamics. It is also true that $p < 0$ states are generally metastable in *thermodynamic* sense, occurring beyond full phase equilibrium. (In this paper we emphasize the difference between the *dynamical* pressure P and thermodynamical one p; the difference will be clarified in Sect. 2.) Still, if there is a potential barrier against the formation of the new phase, such states can be reached in a process, and thermodynamical formalisms based on *local equilibrium* are applicable. Third, temperature and pressure are both thermodynamical intensives, so one would first expect similar types of lack of stability at their negativities. Finally, it is an experimental fact that even $T < 0$ states, called above "completely unstable" can be maintained in laboratory for seconds or minutes [4].

The above list of problems suggests that the behaviour of $p < 0$ states is not without interest. And, since there is consensus about *hydrodynamical* instability, our goal in this paper is to discuss the *thermodynamics* of $p < 0$ states. Our question is: do the fundamental laws and principles of thermodynamics classify $p < 0$ states as something basically different from $p > 0$ ones?

2. *Some $p < 0$ states in physics*

In the Introduction three examples were mentioned. Here we recapitulate them very briefly, and then discuss if these states can be reached in real processes.

For later use we need some basic thermodynamic formulae. Throughout this paper it will be sufficient to consider a matter of three independent thermodynamic extensives, which are chosen as volume V, (internal) energy E and particle number N. (There will be cases *without* one of them). Then the proper potential is the entropy S:

$$S = S(V, E, N). \tag{2.1}$$

From homogeneous linearity

$$S = Vs(e \equiv E/V, n \equiv N/V). \tag{2.2}$$

Entropic intensives are defined as

$$1/T \equiv \partial S/\partial E = \partial s/\partial e, \tag{2.3}$$

$$-\mu/T \equiv \partial S/\partial N = \partial s/\partial n, \tag{2.4}$$

$$p/T \equiv \partial S/\partial V = s - e/T + \mu n/T, \tag{2.5}$$

where T is the temperature, μ is the chemical potential and p is the thermodynamic pressure. Hence "dynamical" pressure P, which can be read off the energy-momentum tensor, cannot be equal with the "thermodynamic" pressure p, but rather p is its "leading part", depending only on thermodynamic variables. The best known deviation comes from viscosity; for more details see Ref. [5]. Henceforth we denote partial derivatives by comma as $\partial s/\partial n \equiv s,_n$. By means of repeated Legendre transformations one can arrive at another potential $\Omega \equiv pV/T$, where $p = p(T, \mu)$, and then

$$s = p,_T, \tag{2.6}$$

$$n = p,_\mu, \tag{2.7}$$

$$e = Ts + \mu n - p. \tag{2.8}$$

It is interesting that quantities of the thermodynamic limit can be used to calculate fluctuations of finite subsystems as well [6, 7]. Without going into details, one can measure fluctuation distances by the metric tensor g_{ik}

$$g_{ik} = -s,_{ik} \tag{2.9}$$

and then the average fluctuations, if small, in a subsystem of volume V^* are obtained as

$$\langle \delta x^i \delta x^k \rangle = g^{ik}/V^*. \tag{2.10}$$

If g_{ik} is positive definite, then the expectation values of extensives remain small in a sufficiently large subsystem. Therefore for small disturbances the system returns to its initial state. Then, according to a mechanical analogy, one may call it *stable* in the absolute ground state and *metastable* if there is another state with higher entropy at the same values of all extensives in a closed system. In the second case large fluctuations may trigger a transition into the other state, but the time scale of such a transition may be much longer than that of the elementary processes. A demonstrative example is the possibility that strange quark matter may be the true ground state of familiar nuclear matter, but, being the transition a high order weak process, still nuclear matter is practically stable for superastronomical times [8].

Now we have all the necessary tools of discussion; let us start.

a) *Overdilute cool nuclear matter*

For approximate equations of state see Refs. [2] and [9]. There is a liquid-gas phase transition below half normal nuclear density [9]. Negative pressures can be reached by diluting nuclear matter well behind phase equilibrium, *if* T is several MeV or less.

This is so because hydrodynamical models of $T = 0$ nuclei use $p = 0$ at normal nuclear density n_0, which is a local compressional energy minimum as well:

$$e \approx m_n c^2 n + (K/18)(n - n_0)^2. \tag{2.11}$$

Then $\mu = m_n c^2 + (K/9)(n - n_0)$ and from Eq. (2.8) for $T = 0$

$$p \approx (K/18)(n^2 - n_0^2). \tag{2.12}$$

This is indeed negative below n_0.

Dynamical calculations of heavy ion collisions show that the matter sometimes enters the $p < 0$ region in the expansion stage [2, 10]. Then the detectable fragments are created at negative pressures, so in principle one could check from fragment distributions if such states existed indeed.

b) *Dilute cool quark plasma*

For quark-gluon plasma remember that in the perturbative regime of QCD for three colours and two light flavours [11]

$$p \approx (\hbar c)^{-3}\{(37\pi^2/90)T^4 + \mu^2 T + \mu^4/2\pi^2 - B^4\}. \tag{2.13}$$

Here B is a characteristic energy of QCD, whose value can be deduced from hadron spectroscopy [12].

This "perturbative" p is negative if both μ and T are sufficiently low, while perturbative treatment is appropriate for sufficiently high density and/or temperature. But these two domains may overlap. For the reasonable $B = 235$ MeV [12] rough approximations [13] yield $p = 0$ slightly above $6n_0$ density, which is moderately high, so then below one may believe in $p < 0$.

However no clear situation is known where expanding quark plasma would enter this region. Dynamical calculations suggest rehadronization near the $p = 0$ line [14].

c) *Cold or tepid symmetric phase of Grand Unification*

Grand Unification [15] has a spontaneous symmetry breaking at $T \sim (10^{14} - 10^{15})$ GeV. Before and after the equations of state can be roughly approximated as [16]

$$p_{\text{sym}} \approx (\hbar c)^{-3}(AT^4 - B^4), \tag{2.14}$$

$$p_{\text{asym}} \approx (\hbar c)^{-3}CT^4, \tag{2.15}$$

where A and C are number constants, proportional to numbers of helicity states of light particles, $A > C$, and $B \sim 10^{15}$ GeV. Then the phase equilibrium temperature is $\sim B$, and $p < 0$ if the symmetric phase is substantially supercooled.

Such energy densities probably existed in the very early Universe. Now, at sufficiently negative dynamical pressures the expansion of the Universe becomes nearly exponential [17]. Extrapolating back isentropically from present observed data, at Planck temperature the scale factor of the Universe turns out to have been cca. 10^{30} Planck radius, which is a completely arbitrary and unexplainable initial condition. (Another aspect of this is that initial conditions must have been fine tuned to 60 digits [17].) If one is reluctant to accept *this*, the alternative is a stage of exponential expansion. Either the matter cools and there will be a subsequent entropy producing reheating [17], or there is a roughly isothermal tepid or hot state [18, 5]; the key point is the necessity of $P < 0$ *and* entropy production. So the present data are strong suggestions for $P < 0$ in the past. Hence $p < 0$ does not follow in the *hot* case (one may get $P < 0$ by viscous stresses [5, 19]); however it does in cold and tepid models.

It seems as if the $p < 0$ states were consequences of completely different mechanisms in the listed cases. However, from thermodynamic viewpoint, Cases a–c) have much common.

3. *The analogy of $T < 0$*

It may be useful to make here a brief detour to see what are the negative *temperature* states like. Following the idea of [20], consider a system of N subsystems, with two states each (N is fixed). The "down" state has $E_i = 0$, the "up" has $E_i = E_0$, $1 \leqslant i \leqslant N$. (One may think of spins fixed in a lattice). Then in a state of energy $E = kE_0$ $N-k$ subsystems are in state "down". Therefore, from the number of microstates

$$S = \ln \{N!/k!(N-k)!\}, \quad k \equiv E/E_0. \tag{3.1}$$

Obviously E has a maximal possible value NE_0 and from Eq. (3.1)

$$S(0) = S(NE_0) = 0. \tag{3.2}$$

Then, via Eq. (2.2) the temperature can be approximately evaluated by Stirling formula,

$$1/T = (1/E_0) \ln \{(NE_0 - E)/E\} + (2E - NE_0)/\{2E(NE_0 - E)\} \tag{3.3}$$

and T vanishes in both limits. S has a maximum at $E = NE_0/2$, there $1/T$ is *0*, and behind it becomes negative.

These states possess negative temperatures; because heat flows to places of higher $1/T$, such states are hotter than $T > 0$ ones. The system is overfilled by energy in some sense and wants to get rid of it. However, in *thermodynamic* sense (at fixed values of extensives including energy E, in a closed system) the state still may be stable, and in this simple model it *is*. Indeed the $S(E)$ curve is convex there, then the metric tensor (2.9) is positive definite, and, furthermore, there is no other competiting state for that energy $> NE_0/2$ there. This is the reason that such states can be maintained for macroscopic time [4].

4. *Thermodynamic pressure and its determination*

Now, p is known if the function $S(V, E, N)$ is given. This $p(e, n)$ must be compatible with information from mechanics. Thus one has to assume S functions, and then must select that one which gives good pressure.

There is an axiomatic construction of thermodynamics, postulating the properties of entropy functions; the Postulates were given by Callen [21]. If thermodynamics has any objection against $p < 0$ states or at least wants to handle differently them, then it must be reflected in connection to a Postulate (contained by either this Postulate System or by an extension of it). Unfortunately, for later use, we are forced to strictly cite the Postulates:
Postulate I: There exist particular states (called equilibrium states) of simple systems that, macroscopically, are characterized completely by the internal energy U, the volume V, and the mole numbers N_1. $N_2, \ldots, N_r$ of chemical components (extensive parameters).
Postulate II: There exists a function (called the entropy S) of the extensive parameters of any composite system, defined for all equilibrium states and having the following property. The values assumed by the extensive parameters in the absence of an internal constraint are those that maximize the entropy over the manifold of constrained equilibrium states.

Postulate III: The entropy of a composite system is additive over the constituent subsystems. The entropy is continuous and differentiable and is a monotonously increasing function of the energy.

Postulate IV: The entropy of any system vanishes in the state for which $\partial U/\partial S = 0$, that is, at the zero of temperature. (Note that in our paper E plays the role of U). Any function satisfying these Postulates is a legal entropy function. The first sentence of Postulate III is the homogeneity of order 1 and Postulate IV is the Third Law.

It is easy to see that these Postulates do not require anything directly for p. Namely

$$p = S_{,V}/S_{,E} \tag{4.1}$$

and no Postulate states anything particularly for the V dependence. So, within Callen Postulates, $p < 0$ and $p > 0$ states do not belong into different classes. Quite oppositely, $T < 0$ states contradict to the last half sentence of Postulate III. This condition could be lifted, but that is not the object of this paper.

Now, Callen Postulates enable us to determine the $S(V, E, N)$ function of a particular new system via Postulates I and II, by establishing equilibria between a whole series of old, known systems (at least 3 different ones) and the new one, and measuring the extensives on both sides [22, 23]. Then the S function of the new system will be unique. However, there is a freedom to simultaneously change the entropies of *all* systems as

$$S_\alpha(V, E, N) \rightarrow K^2 S_\alpha(V, E, N) + AV + BE + CN, \tag{4.2}$$

where K, A, B and C are universal constants, the same for each system [23], because these zero point shifs and rescalings of entropic intensives do not affect the equilibrium of any pair. These changes are conform with all Postulates, save that the last condition of Postulate III imposes a *negative* or zero lower bound on B. Observe that the H-theorem of statistical physics does not help (even for systems having a clear statistical desription); the additive terms in S correspond to terms additive in the H-function, keeping its definite production because of conservation laws.

However, clearly, if there is no further constraint on A, there cannot be on the sign of p, either: introducing a large negative A p becomes negative even for familiar states. Of course, our p should be "as similar as possible" to P measured in mechanics, and this may define a proper A, at which thermodynamics and mechanics are compatible (if they are, indeed), but negative pressure states were concluded just from some kind of mechanics or dynamics.

So the moral of the present Section is that the original Callen Postulates do not put a lower boundary for pressure.

5. The possibility of further constraints

However, still this fact might be considered as a deficiency of Callen Postulates. By other words, if one is deeply convinced that $p < 0$ states do not belong to regular thermodynamics, he may rule them out by means of additional Postulates. The simplest such one is

Proposition: First at the end of Postulate III be added "... and the volume".

Then $p > 0$. Now the Postulates exclude negative pressures as well as negative temperatures. However, we are not yet ready: freedom (4.2) now may contradict to the Postulates. The zero point of p/T is to be fixed.

For doing this the zero point of $1/T$ is a good analogy. A subgroup of freedom (4.2) was first observed by Guggenheim, who, however, wanted to make the entropy unique. To achieve this he introduced the notion of quasistatic adiabatic processes into the Postulates, and formulated a fifth one in terms of such processes. But then the postulate system becomes a mixture of Gibbs and CKC constructions, hard to match together. In order to avoid this one may choose Postulate V very similar in structure to Postulate IV [23]:

Proposition:

Postulate V:

$$\lim_{E\to\infty} \partial S/\partial E = 0.$$

(Such limits are, of course, meant by keeping the other extensives fixed and finite). Then, indeed, the 0 point of $1/T$ is fixed, because this condition (if can hold at all simultaneously for a whole set of systems), can hold only with a unique value of B. This Postulate expresses some feeling that putting more and more energy into a system a universal asymptotic state is being approached. Now, zero point fixed, Postulate III rules out negative temperature states. (By this, we do not state that it is advisable to rule them out).

Then one may apply the same idea on pressure. Postulate III has been appended, and now let there be

Proposition:

Postulate VI:

$$\lim_{V\to\infty} \partial S/\partial V = 0.$$

This proposition puts the zero point of pressure to asymptotically diluted states, which seems natural enough. Namely, consider a system of finite energy and particle content. If the volume of the system is growing without change of E and N, any unit volume goes closer and closer to some kind of vacuum, and the Postulate expresses that these asymptotic states of any system do not disturb each other in contact by volume competition.

Then the extended postulate system (whose applicability has not yet been investigated) rules out both $T < 0$ and $p < 0$ states. This would be something in the spirit of § 12 of Ref. [1], although now $p < 0$ and $T < 0$ states are *similarly* forbidden (it is difficult to do otherwise in thermodynamics). However, as will be immediately seen, this Postulate System excludes existing material *systems* (not simply states) from thermodynamic treatment.

6. *Special systems*

We may have two kinds of problems with the additional Postulates V and VI. Either they do not hold in Nature or they cannot be imposed. The first problem is not so serious and definitely not explicit. Take two different material systems and put infinite energy into both without changing V and N. The experiment cannot really be done; however, by investigating limiting processes one at least does not have evidence against possibility of limiting thermal equilibria. Similarly, we have no counterevidence for the belief that two systems of finite E and N will be in mechanical equilibrium when both V's go to ∞.

However, in Sect. 3 we saw a system where the limit $N = \mathit{fixed}$, $E \to \infty$ *did not exist*. Such systems are called *special* [24]. Similarly, in Cases b–c) in Sect. 2 one cannot perform the limit: E is fixed, N is fixed, $V \to \infty$. In order to see this, consider Case c). From Eqs (2.6–8), (2.14) one gets

$$e = (\hbar c)^{-3}(AT^4/3+B^4). \tag{6.1}$$

Therefore energy density has an absolute minimum, consequently with $V \to \infty$ $E \to \infty$ as well. If there is a positive lower bound for energy/volume, the system cannot reach asymptotic state in volume at fixed energy, just as a system of finite upper bound cannot reach asymptotic state in energy at fixed volume. Since we have seen how and why the second case leads to a region near to $E_{\max}$ where $S_{,E} < 0$, it is no surprise that in the first case there is a region near $V_{\max}$ with $S_{,V} < 0$. This is the situation in Cases b) and c).

Case a) is different. There is a state in complete equilibrium with vacuum: the nuclear matter in normal state. There the energy density has a *local* minimum. The limit of Postulate VI does exist, and yields a zero of pressure compatible with any experience. (Infinitely dilute nuclear matter keeps equilibrium with infinitely dilute other matters.) However between $n = n_0$ and $n = 0$ the energy density is above 0, so dilution must take the needed energy from heat, therefore there S is *decreasing* function of V. If any *well founded* theory predicts this behaviour of the energy density, then p must be negative there.

Consequently: the original version of Callen Postulates excludes $T < 0$ states (which exist) but tells nothing against $p < 0$ ones. By an extended postulate system $p < 0$ states can be excluded too, but then some systems have no thermodynamic description at all. If one accepts Postulates V and VI for all systems *for which the limit exists*, and for other, special, systems the zero points are deduced from equilibrium measurements, and, simultaneously, he removes the requirement of monotonous behaviour from Postulate III, then all systems can be described, and they are classified into two classes. In the special or exceptional class (for which at least one limit does not exist) one can find all the known kinds of systems having $T < 0$ states, and some systems able to produce $p < 0$ as well, *but not* the nuclear matter or the van der Waals gas (the only realistic example mentioned in Ref. [1]). Furthermore, the Postulates (any variant mentioned here) classify systems and not states. So we have been unsuccessful in finding fundamental thermodynamic difference between *all* $p > 0$ and *all* $p < 0$ *states*.

7. *Metastability*

Still *metastability* of $p < 0$ states is indisputable fact in all cases mentioned here, and this deserves some discussion. For cool dilute nuclear matter Danielewicz found [2] that the $p < 0$ region can be divided into two parts. At higher densities the local state is metastable, between a critical density and 0 there is instability against arbitrarily small fluctuations. For QCD the perturbative treatment and for GUT the one loop approximation cannot give instability, but we know that the state must become unstable somewhere.

In all three cases the $p < 0$ states occur when (with dilution or cooling) a possible phase transition has not happened. If $p < 0$ states are dominated in entropy by phase mixtures with the same V, E and N in the system, then large fluctuations will carry the system there, and this is a kind of *metastability* mentioned in Ref. [1].

Now one may ask if this metastability is general in $p < 0$ states or not. (The Postulate System is silent in this question). It seems that its generality can be proven at least for systems fulfilling Postulate VI and having $p > 0$ states in the neighbourhood of $n = 0$.

At $n = 0$, $p = 0$ (cf. the Postulate). The stability against small fluctuations requires $\partial p/\partial n > 0$; let us assume first this stability. Then e grows faster than linear everywhere. Since at small n's $p > 0$, it will remain positive always. There remains the case when the stability breaks down somewhere; but then there is possibility for phase equilibrium on the two sides of instability [3]. Since on the low density side $p > 0$, it is so at the high density side too. Then p will grow until the next instability because of $\partial p/\partial n > 0$, where we can repeat the argument. For more details see Ref. [25]. So indeed, it is necessary to go beyond a phase boundary to get $p < 0$ states of familiar systems.

However, it is easy to manufacture a counterexample, although without physical background, for the generality of such metastability by the pressure function at $T = 0$

$$p = -an+bn^2. \tag{7.1}$$

The corresponding energy density is the rather strange

$$e = cn-an\ln(n)+bn^2 \tag{7.2}$$

which, if c is sufficiently positive, is positive everywhere. This system does not have phase transition at all, so now negative pressure states near $n = 0$ cannot be metastable in *thermodynamic* sense.

Even then, the above stability condition tends to act asymmetrically on T and p (or rather on $1/T$ and p/T). But it is easy to understand this asymmetry. Because of the homogeneous linearity of S, one must not form the wanted negative definite matrix from the second derivatives of S with respect *all* the extensives; one must be omitted. This is done by using *densities*, as in Eq. (2.9), but then the there is one intensive, just p/T (cf. Eq. (2.5)) which is not simply a first derivative. Furthermore, V does not have nontrivial density. So, while stability simply imposed $(1/T)_{,E} < 0$, which in itself obviously does not prevent $1/T$ to go into negative, it imposes something structurally different on p.

8. Conclusion

We have investigated, what comes from thermodynamics about the possibility and stability of states of negative pressures. The results are as follows:

Such states may be both stable and unstable against small fluctuations; there are predictions for both behaviours, even in the same system (but in different states). In all known cases these states appear in situations when a first order phase transition is possible, but has not happened. This characteristics seems fairly general. Because of the virtual existence of a higher entropy phase with the same total extensives these $p < 0$ states vanish by large fluctuation (metastability), but the lifetime can be anything, depending on the potential barrier. In contrast, the simplest $T < 0$ system, the spin lattice, is not connected with any phase transition, so $T < 0$ states could remain forever, *if no other degrees of freedom* (e.g. lattice oscillations) *exist*. That is, the *thermodynamic* (*not hydrodynamic*) stability of $T < 0$ states tend to be *greater* than that of $p < 0$ states, which is rather unexpected, but is conform to the remarkable lifetime of negative temperatures in at least some experiments.

Usual thermodynamics does not prohibit $p < 0$. Callen Postulates do not restrict the sign of $S_{,V}$, while excluding $T < 0$. One may extend the postulate system without sctructural changes to exclude $p < 0$ *states*; but then the Postulates exclude all *systems* which have $p < 0$ *states* anywhere, so this extension seems too strict. Furthermore, then there will not be any fundamental difference between $T < 0$ and $p < 0$ states.

The *thermodynamic* reason of negative pressures is quite explicit: it is connected either with a positive lower bound for energy density, when in the limit $E =$ fixed, $V \to \infty$ the heat must be used up, so $\partial S/\partial V < 0$, or with $p = 0$ in a not asymptotically diluted state after which $e > 0$ and again heat is eaten up with further dilution. Therefore we must consider such $p < 0$ states quite natural from thermodynamic viewpoint.

The instability of the *hydrodynamics flow* is another phenomenon having no direct connection with thermodynamics, and appearing on a different time scale. Indeed, there are models in which $p > 0$ but $P < 0$, then even thermodynamic *metastability* does not occur, still the flow may be unstable. But even the generality of this hydrodynamic instability is questionable. In cosmology, the expansion is faster with $p < 0$ than with $p > 0$. If sufficiently large (gravitationally self-governing) parts are in such state, they will relatively eat up vacuum between them, so there will not be instability against droplet formation.

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