

## Thermodynamic Equations of State with Three Defining Constants\*

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**Abstract**—The well-known empiric Van der Waals equation contains two constants characterizing a given gas, so that, in general, it depends on the characteristic interaction of particles of the given gas. In this paper, equations depending on three defining constants. are constructed on the basis of the mathematically rigorous results obtained by the author in recent papers.

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*Key words:* Bose condensate, fractal dimension, number of degrees of freedom, equations of state, theory of imperfect gases, interaction potential of particles, Irving Fisher law.

1. The relationship between the Bose–Einstein distribution and number theory was studied, in particular, in the papers [1], [2], [3], [4]. As an example of a simple model of Bose condensate, consider Koroviev’s trick (well known from Bulgakov’s novel “Master and Margarita”) of scattering money bills in a variety theater’s audience.

According to number theory and Kolmogorov complexity, if Koroviev had one million bills and the number of spectators were ten thousand, then only one thousand of them would get bills. The other nine thousand would not get a single bill (and presumably would die of hunger). This is exactly a model of Bose condensate. But if the spectators united into groups of ten and agreed to divide the bills between themselves, then, figuratively speaking, no one would die, i.e., the number of spectators (of particles) would be preserved. But the association of the spectators into groups would mean a constraint on their degrees of freedom, just as the association of particles into clusters.

This leads to two conclusions.

First, from the mathematical point of view, this example is equivalent to the existence of a two-dimensional Bose condensate, but this refutes the physicists’ postulate (dating back to Einstein) that no two-dimensional condensate does exist. It turns out that, in the Bose–Einstein distribution, it is necessary to add a special term taking into account the fact that the number of particles is finite. Simultaneously, it provides the asymptotic distribution function (previously not known) for the number of bills obtained by groups of spectators.

Second, the Bose condensate can be regarded as an association of dimers, trimers, and clusters, not only of “frozen” particles precipitating to the Bose condensate. And this applies to a classical gas, not a quantum, one. Thus, under the condition  $N = \text{const}$ , the Bose–Einstein distribution can be regarded as a distribution for a classical gas and the degeneracy temperature as the critical temperature [5].

All this has given rise to the development of a new approach to thermodynamics (in which the number of particles is preserved, but the number of degrees of freedom, i.e., the fractal Hausdorff dimension, is variable) and, further, also to economics (in which constraints imposed on trade are a new parameter). The new theory in thermodynamics is in agreement with experimental data to within 3–5% on the average. Near the critical points, this agreement is significantly better than that furnished by computer simulation methods. In thermodynamics, a number of recent experiments concerned with the so-called jamming (incompressibility of glass and fluids) and other anomalies have now been mathematically justified.

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2. Van der Waals wrote his remarkable equation of state with two defining constants  $a$  and  $b$ . However, the Lennard-Jones potential describing the interaction between particles (6–12) and also containing two defining constants does not lead to the Van der Waals equation either in theoretical calculations [6] or in experimental data even for noble (inert) gases.

Following our ideas based on the analogy with economic laws, we shall construct equations of state with reference to *three* fixed points.

First, consider the Irving Fisher economic law for assets

$$PQ = Mv, \quad (1)$$

where  $PQ$  is the amount of merchandise in its money equivalent, ( $Q$  is the merchandise and  $PQ$  is its money equivalent),  $M$  is the money supply, and  $v$  is the velocity of circulation.

Suppose that  $M_0$  is some original money supply. (If the Fisher law is considered historically, then it is expressed via its gold equivalent.) Let us consider constraints on formula (1) related to the overproduction of merchandise in the form of the following statement. There exist constants  $a$  and  $b$  such that, for

$$PQ \geq bM_0 \quad \text{or} \quad v \geq c,$$

there is no money supply  $M$  satisfying relation (1). More precisely, the money supply  $M$  satisfying relation (1) exists only under two conditions:

$$PQ \leq bM_0, \quad v \leq c. \quad (2)$$

The constraint (2) is similar to the restriction on the velocity of particles in mechanics. As is well known, it led to the revision of the Newton–Galileo mechanics.

Now consider the equation of an ideal gas

$$PV = NT, \quad (3)$$

where  $P$  is the pressure,  $V$  is the volume,  $N$  is the number of particles, and  $T$  is the temperature. Suppose that  $N_0$  is the number of particles as  $T \rightarrow 0$ ,  $P \rightarrow 0$ . Then a similar statement is of the form: relation (3) holds only under the condition

$$T \geq c, \quad E = PV \geq bN_0,$$

where  $c$  and  $b$  are some constants.

Relation (3) for an ideal gas can be rewritten as

$$P = Tg, \quad (4)$$

where  $g = N/V$  is the density of the gas. Then our constraint can be written as follows:

*For a given gas, there exist constants  $c$  and  $b$  such that if  $T > c$  or  $P > b$ , then there is no density  $g$  satisfying relation (3).*

This question can be stated differently. Suppose that the pressure  $P$  and the temperature  $T$  are given. Does there exist a density  $g$  satisfying relation (4) for a given imperfect gas (or a mixture of gases)?

In modern theory of imperfect gases, it is usual to consider the plane  $\rho, T$  and, in this plane, condition (3) corresponds to the so-called Zeno-line, which, as given by experiment, is a segment of the straight line

$$g = g_0 \left(1 - \frac{T}{T_B}\right), \quad T < T_B, \quad (5)$$

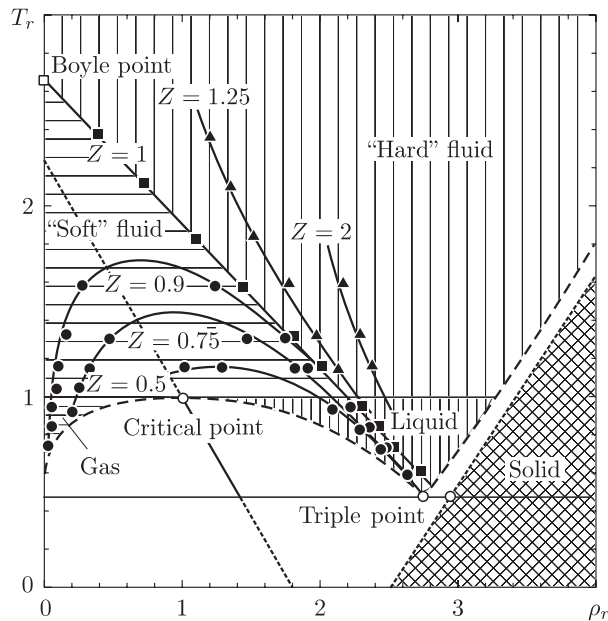
where  $g_0, T_B$  are constants (the constant  $T_B$  is called the “Boyle temperature”).

This dependence was first noticed by Bachinskii, who justified it empirically. Let us present the modern general scheme for an imperfect gas in the plane  $T, \rho$ .

As was already pointed out by the author [7], the thermodynamic equations of state containing the quantities:  $P$ , the pressure,  $T$ , the temperature,  $\mu$ , the chemical potential and, respectively,  $V$ , the volume,  $S$ , the entropy,  $N$ , the number of particles, constitute a three-dimensional Lagrangian manifold in the six-dimensional phase space; moreover,  $P, T, \mu$  play the role of coordinates, while  $V, S, N$ , respectively, play the role of the corresponding momenta.

Therefore, for a fixed number of particles  $N$ , the diagram on the plane  $\rho = N/V$ ,  $T$  given in Fig. 1 is the projection on one of the planes of the phase space. Under such a projection, focal and caustic points appear.

We shall see below that the passage of “soft fluids” to “hard fluids” occurs on the Legendre manifold in the five-dimensional tangent space *continuously* without phase transitions of the first kind. Disregard for these phenomena may lead to catastrophic errors, such as those occurring in the attempts by British Petroleum engineers to stop the oil leak in the Gulf of Mexico; namely, “hard fluids” were formed and they prevented the initial attempts to succeed.



**Fig. 1.**  $T$ – $\rho$  diagram for gases corresponding to simple liquids,  $T_r = T/T_{cr}$ ,  $\rho_r = \rho/\rho_{cr}$ . The  $Z = \frac{PV}{NT} = 1.0$  is a line (Zeno-line) on the phase diagram. For states with  $Z > 1.0$  (hard fluids) repulsive forces dominate. For states where  $Z < 1.0$  (soft fluids) attractive forces dominate.

The slanting line issuing from the Boyle point in Fig. 1 is called the Zeno-line and is of the form (5).

In studying the pressure as a function of the density  $\rho$ , an ambiguity arises on the Zeno-line:

$$P = c\rho \left( 1 - \frac{c\rho}{4b} \right). \quad (6)$$

Hence the quantity

$$\rho = \frac{2b}{c} \pm \frac{1}{c} \sqrt{4b^2 - 4bP} \quad (7)$$

takes two values for  $P < b$  and becomes complex for  $P > b$ , a typical simple caustic (or the turning point in the one-dimensional quantum Schrödinger equation).

This means that Fig. 1 provides very scanty information about the thermodynamic equations of state. But it is important that Fig. 1 contains the key points indicated above as well as the critical temperatures. For the third point, we shall take the quantity

$$Z_{cr} = \frac{P_{cr}V_{cr}}{T_{cr}R},$$

where  $R$  is the gas constant.

It is well known that, for gases corresponding to simple liquids, the linearity of the Zeno-line is in good agreement with experimental data. Water is not a simple liquid. Let us present a diagram for water in Fig. 2.

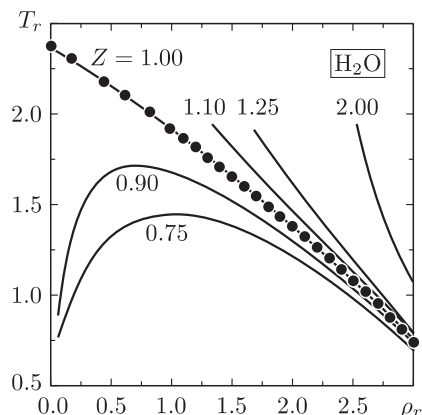


Fig. 2.  $T$ - $\rho$  diagram for water.

Here the Zeno-line deviates from a straight line. However, it is seen from the other curves  $Z = \text{const}$  that there exists a line separating “hard fluids” and “soft fluids”. However, for it, the Boyle temperature is somewhat greater than 2.5 (the definition of the Boyle point given by the author [6]) differs from the one universally used.

3. The dressed or “thermal” potential  $\varphi(r)$  is attractive. In addition, because the volume  $V$  is a large parameter, it follows that if

$$\varphi(r) = U\left(\frac{r}{\sqrt[3]{V}}\right)$$

is expanded in terms of  $1/\sqrt[3]{V}$ , then

$$U\left(\frac{r}{\sqrt[3]{V}}\right) = C_1 + \frac{C_2 r}{\sqrt[3]{V}} + \frac{C_3 r^2}{(\sqrt[3]{V})^2} + O\left(\frac{1}{(\sqrt[3]{V})^3}\right). \quad (8)$$

Expanding

$$C_1 + r^2 = \frac{(r - r_0)^2}{2} + \frac{(r + r_0)^2}{2}, \quad (9)$$

where  $C_1 = r_0^2$ , we can, just as in [8], separate the variables in the two-particle problem and obtain the scattering problem for pairs of particles and the problem of their cooperative motion for  $r_1 + r_2$ .

Then, in the scattering problem, an attractive quadratic potential (inverted parabola) is added to the Lennard-Jones interaction potential.

For this problem, we can find, just as in [8], a point corresponding to the temperature at which the well capturing the dimers vanishes, and thus determine the Zeno-line.

In fact, this procedure is equivalent to the procedure of virial expansion, which was first proposed by Bachinskii at the beginning of the last century.

For mathematicians, the proposed method, is, possibly, less heuristic and clear-cut than that of virial expansion with the use of the model Van der Waals equation. As we shall see in what follows, the Zeno-line resulting from rigorous distributions is not even a smooth curve: it has a slight break, a phase transition of the second kind. A similar break was experimentally discovered by Holler in the mid 1960s. It is very difficult to discover such a break, because a continuous curve can always be constructed from experimental points.

It should be noted that the relevant experiments were carried out on the surface of the earth, and hence were subjected to gravitational pull. Therefore, the coagulating drops fell to the ground and the

liquid was underneath and the gas above. In the other cases, experiments were conducted in narrow test-tubes and films were formed between the walls of the test-tubes. And this meant that the wetting of walls, their attraction or repulsion played a significant role.

In [6], we characterized the formation of “three-dimensional” clusters as a characteristic of the phase transition “gas–liquid.”

Here, however, many questions arise. The notion of “first coordination sphere,” i.e., of the number of molecules of a liquid enclosing a given molecule significantly depends on temperature, pressure, and depth so that it is difficult to determine the dimensions of a three-dimensional cluster as a drop nucleus. Therefore, experimental data may have significant discrepancies near the critical point.

4. Consider the potential  $\Omega_\gamma$  for the Bose–Einstein distribution for  $\gamma = (d - 1)/2$ , where  $d$  is the fractal dimension.

For the dimension  $d = 2\gamma + 2$ , the potential  $\Omega_\gamma$ ,

$$\Omega_\gamma^{\text{id}} = \frac{\pi^{1+\gamma} T^{2+\gamma} V}{\Gamma(2+\gamma)} \int_0^\infty \xi^{1+\gamma} \left\{ \frac{1}{e^{(\xi-\kappa)} - 1} - \frac{N}{e^{(\xi-\kappa)N} - 1} \right\} d\xi, \quad \kappa = \frac{\mu}{T}, \quad (10)$$

is of the form

$$P = \frac{\partial \Omega_\gamma^{\text{id}}}{\partial V}, \quad N = \frac{\partial \Omega_\gamma^{\text{id}}}{\partial \mu}, \quad S = \frac{\partial \Omega_\gamma^{\text{id}}}{\partial T}. \quad (11)$$

For  $\gamma > 0$ , the second term in (10) can be dropped (it tends to zero as  $N \rightarrow \infty$ ).

In particular,

$$S = \frac{\partial \Omega_\gamma^{\text{id}}}{\partial T} = N((\gamma + 2)Z(\gamma, \kappa) - \kappa). \quad (12)$$

Suppose that  $\gamma = \gamma_0$  is the point at which the condensate appears. On the isochore issuing from the condensate point  $T_r = 1$ ,  $P_r = 1$ , for which  $\kappa = 0$ , in view of the increasing entropy with  $\gamma$  for  $\gamma \leq \gamma_0$ , a phase transition of the second kind (a sharp break; see Fig. 3) occurs at the point  $\gamma = \gamma_0$ ; further, for  $\gamma = \gamma_0$ , the isochore changes from  $\kappa = 0$  to  $\kappa = -\infty$  [9].

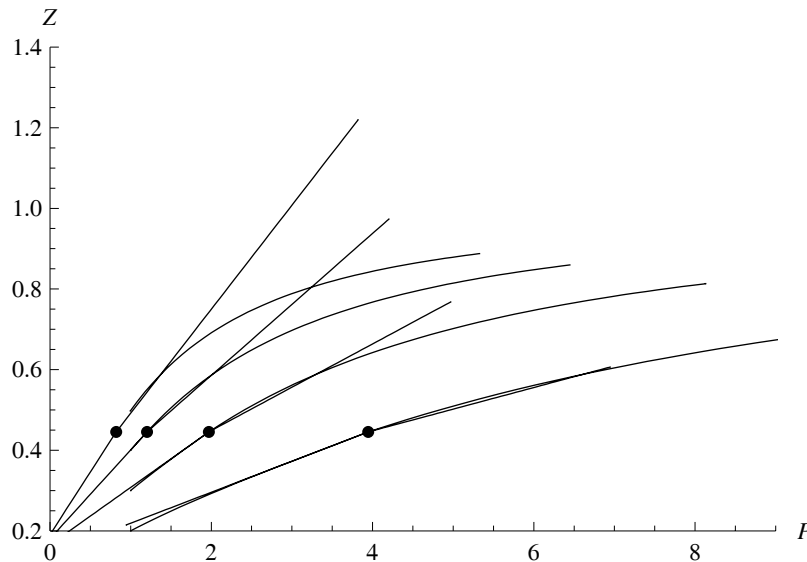


Fig. 3. Breaks on theoretical isochores for  $Z_{\text{cr}} = 0.45$ .

First, consider the generalization of the potential  $\Omega$ , which corresponds to the fractal dimension  $d = 2\gamma + 2$ . Let us set

$$V_r = \arcsin\left(\frac{\tilde{V}}{r}\right). \quad (13)$$

Hence

$$\tilde{V} = r \sin(V_r), \quad \tilde{P} = r \cos(V_r).$$

For  $\tilde{Z}$ , we obtain

$$\tilde{Z} = r \frac{\tilde{V}\tilde{P}}{T}. \quad (14)$$

For  $r < 1$ , this describes a compression transformation along the coordinate  $Z$ . This is a natural transformation of a classical ideal gas of dimension  $d = 2\gamma$  in view of the fact that it is not known how the fractal dimension  $d$  is related to the real volume, which is the convex hull of the fractal (see [9]).

For the potential  $\Omega$ , let us perform the rotation transformation about the point  $V_\gamma/R$  corresponding to the point at which the condensate appears and to  $P = 1$ ; see Fig. 4.

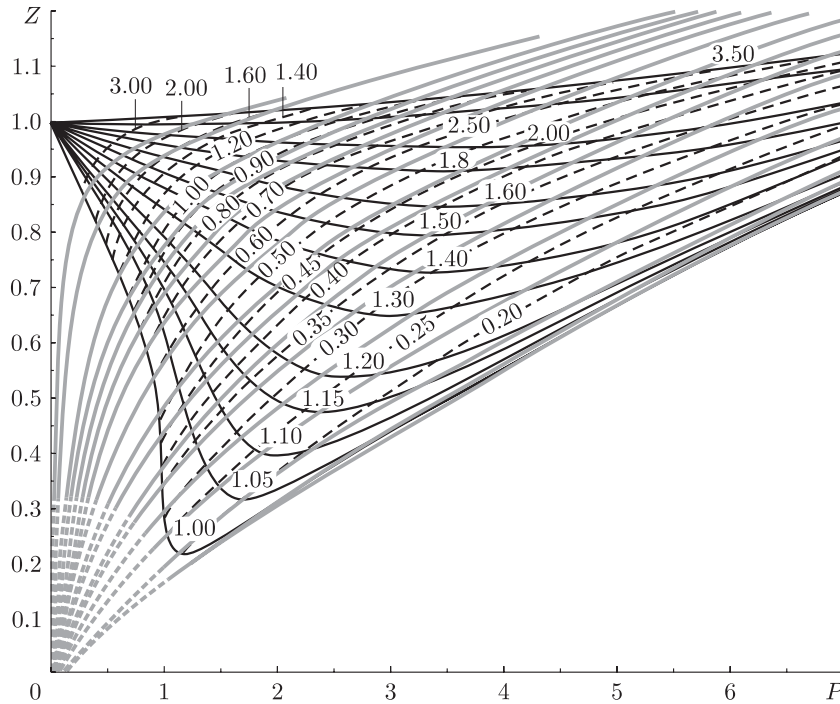


Fig. 4. Theoretical isochores under rotation.

In Fig. 4, the dotted lines below the vertical isotherm  $T = 1.00$  show the continuations of the theoretical isochores for  $\mu = 0$  and decreasing  $\gamma$ . The dotted lines with values  $V/R = 0.50, 0.45$ , etc., indicate experimental isochores. If we perform the compression transformation with respect to  $Z$ , then the theoretical isochores will bend and almost coincide with the experimental ones. But, in that case, we will not see the experimental dotted lines.

Let us now choose the angle of rotation and the transformation (14) so that after the rotation for  $Z = 1$  there will be exact coincidence with the points

$$\tilde{P} = c \frac{R}{V} \left( 1 - \frac{cR}{4bV} \right)$$

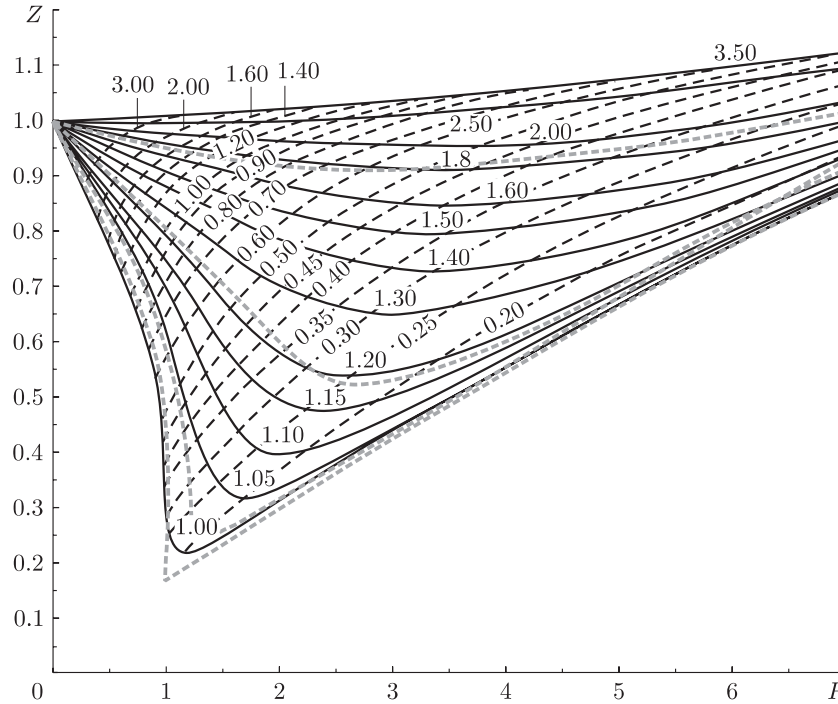
for  $\rho = R/V_\gamma$  and  $\rho = 1$ ; for  $\rho < 1$ , we continue the curve  $Z = 1$ , considering the gas as ideal, and all the isotherms, respectively, we continue by straight-line segments reaching the point  $\tilde{Z} = 1$ ,  $\tilde{P} = 0$  (similarity transformation).

For

$$\tilde{Z} < Z_\gamma = \frac{V_\gamma}{R},$$

using the previous transformation (13), (14), we perform a rotation for each  $\tilde{Z} = \tilde{V}/R$ ,  $\tilde{P} = 1$  so as to reach, along  $\tilde{Z} = 1$ , the point  $\tilde{P}$  corresponding to the Zeno-line.

It turns out that even if we do not perform the transformation (13), (14), the agreement with the experimental isotherms is rather good (see Fig. 5). Choosing the transformation (13), (14), we can significantly improve this agreement.



**Fig. 5.** Isotherms for  $T_r > 1$ . The bold lines denote theoretical isotherms in which only rotation is taken into account.

Let us consider the rotation transformation and the change of the potential  $\Omega$ .

$$P' = 1 + \alpha P + \beta V, \quad V' = -\beta P + \alpha V, \quad \alpha^2 + \beta^2 = 1.$$

We obtain the potential  $\alpha\Psi = V' dP'$ . Hence, substituting  $V'$  and  $P'$ , we obtain

$$(-\beta P + \alpha V)(\alpha P + \beta V) = V dP + d\left\{\frac{\alpha\beta}{2}(V^2 - P^2) - \beta^2 PV\right\},$$

$$\Psi = \Omega + \frac{\alpha\beta}{2}(V^2 - P^2) - \beta^2 PV$$

or

$$\Psi = \Omega(P', V') + \frac{\alpha\beta}{2}(V'^2 - P'^2) - \beta^2 P'V',$$

$$[-\beta P + \beta + (\alpha V - 0.3\alpha)](\alpha dP + \beta dV) = \frac{\alpha\beta}{2}(V^2 - P^2) - \beta^2 PV + \alpha\beta P - 0.3\alpha\beta V.$$

Below the condensate point, for  $P = 1$ , the angle of rotation depends on  $V_\gamma/R$  for  $\gamma < \gamma_0$ . This is a fairly complicated transformation. Nevertheless, since it occurs in the plane  $\tilde{P}, \tilde{V}/R$ , it does not involve the coordinates  $T, S/R$  and preserves the Lagrange property, because, in the two-dimensional phase space, any smooth transformation preserves the Lagrange property and the coordinates  $T, S/R$  remain unchanged. It is important that  $S/R$  is still the logarithm of the number of possible variants.

The physical meaning of the rotation consists in the following. A rotation through  $90^\circ$  indicates the passage of the thermodynamic potential to free energy (see the explanation of the physical meaning in [10]). A rotation through a smaller angle means that we consider some “mean” potential between the

free energy and the thermodynamic potential, where the usual imperfect specific volume is some variable quantity depending on the fractal dimension, while the dimers and clusters are created at some points in space and vanish at other points. Only in the total volume does the number of particles remain constant. The fractal dimension is preserved, as well as the number of degrees of freedom, in the whole volume, not in its isolated parts. This process is especially obvious on the line  $\tilde{P} = 1$ , where the real volume and rotation are related to each other, while the real volume is directly related to the fractal dimension.

Let us now pass to the projection of the two-dimensional manifold (obtained above) in the six-dimensional space on the four-dimensional phase space. The equality  $N = \text{const}$  cuts a two-dimensional surface out of the three-dimensional Lagrangian manifold. The following group property holds: as  $N$  is changed  $k$ -fold, so are the quantities  $V$  and  $S$ . This implies that this manifold is cylindrical. Therefore, it can be projected along  $\mu$  and  $N$  from the six-dimensional phase space on the four-dimensional space, where the explicit action determining the Lagrangian manifold is the Gibbs potential  $\Phi = N\mu(P, T)$ .

Since we have  $N = \text{const} = R$ , this equality cuts out a two-dimensional cylindrical (by the group property) manifold depending on the parameter  $d = 2\gamma + 2$ . Projecting it along  $N$  and  $\mu$ , we obtain a two-dimensional manifold in the four-dimensional phase space  $P, V/R, T, S/R$ . First, consider this projection using  $\Omega_\gamma^{\text{id}}$  as an example.

Here  $P, T$  play the role of coordinates, while  $V/R, S/R$  that of momenta. Using the relation  $P = \partial\Omega/\partial V$ , we obtain

$$P = T^{\gamma+2} \int_0^\infty \frac{\varepsilon^{\gamma+1} d\varepsilon}{e^{-\mu/T} e^\varepsilon - 1} \quad \text{for } \gamma > 0 \quad (15)$$

and

$$P = T^{\gamma+2} \int_0^\infty \xi^{1+\gamma} \left\{ \frac{1}{e^{(\xi-\kappa)} - 1} - \frac{N}{e^{(\xi-\kappa)N} - 1} \right\} d\xi \quad \text{for } 1 < \gamma \leq 0. \quad (16)$$

From (15), expressing  $\mu$  as a function of  $P$  and  $T$ :  $\mu = \mu(P, T)$ , we obtain the potential

$$\Phi = R\mu(P, T)$$

and the relations

$$V = \frac{\partial\Phi}{\partial P}, \quad S = \frac{\partial\Phi}{\partial T}.$$

This gives rise to a potential of the type of the Gibbs potential  $R\mu(P, T)$  determining the Lagrangian manifold nonuniquely projected onto the plane  $P, T$ .

The usual rule is used for the selection of points at which different branches of the projections are joined. The “quantization” of thermodynamics is carried out by the tunnel canonical operator, as was described in [11].

Just as above, we can project the Lagrangian manifold (after the rotation) in transformed (with respect to  $V$  and  $P$ ) coordinates along  $\mu$  and  $N$  on the four-dimensional phase space. Since the projection does not depend on the order of transformation of the coordinates  $P$  and  $V$ , it follows that the same transformation can be performed after the projection.

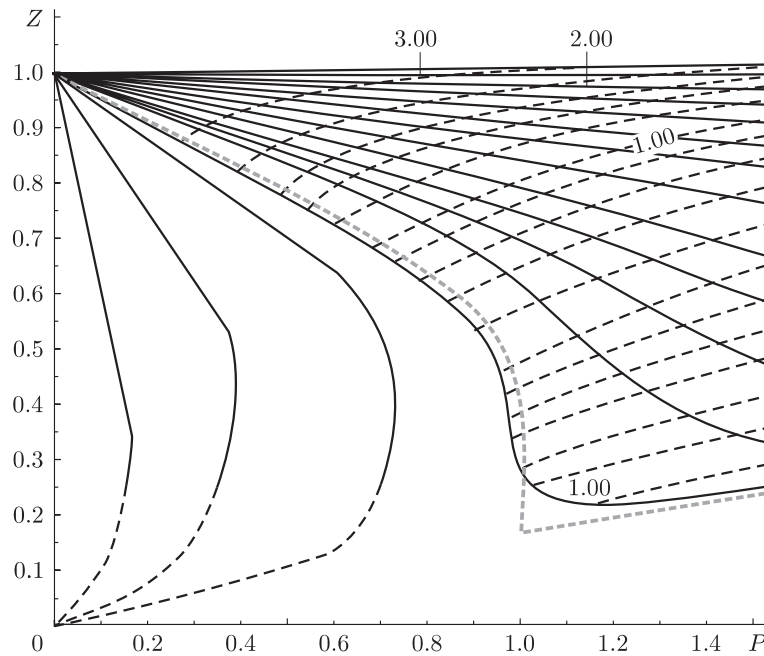
The fact that the potential “mixed” from the free energy and the thermodynamic potential arises can easily be explained; see the heuristic example given in [6]. The phase transition in liquids is treated there as the creation of “three-dimensional” clusters or domains, as the author called them, in which there exists a particle surrounded on all sides by other particles (constituting a small coordination sphere).

This transition can be observed as a passage from protuberances (critical opalescence) to a totally blurred medium of liquid drops inside which the pressure is higher than in the enveloping gas because of “surface tension.”

There is a well-known experiment in which a small drop is joined by a capillary to a big one and it gradually diminished owing to the higher pressure in the small drop.

In an equilibrium state, the liquid will have the form of a spherical drop; its surface tension determines the pressure jump. The change of the angles between the specific volume and pressure is responsible for these phenomena and explains them.





**Fig. 6.** Isotherms for  $T_r < 1$  determining the phase transition “gas–liquid.” On the isotherms  $T_r = 0.9, 0.7, 0.5$ , the dotted line indicates the liquid phase.

Using the isochores from Fig. 3, we can construct isotherms for  $T < T_{cr}$ .

In Fig. 6, the “rotated” pressure  $P$  already takes into account the effect of surface tension. Since the density in a vessel where the transition to the critical state occurs, is equal to  $10/3$ , it follows that the points on the isotherm corresponding to the transition “gas–liquid” are uniquely determined from the condition that the total density (“weight”) is equal to  $10/3$ .

Thus, pressure, together with volume, undergoes a phase transition contrary to the notions of standard thermodynamics. These notions are only approximate with respect to certain small parameters, just as all notions discussed in [12].

**Remark 1.** The fractal dimension corresponding to  $Z = 0.3$  does not, in general, remain constant as  $P$  tends to zero; the number of clusters decreases, and hence the dimension tends to 3.

**Remark 2.** The angles of rotation  $\alpha$  (in radians) in the plane  $P, V$  depending on  $V$  are given in the following table.

$$V \geq 0.3 \quad \alpha = 0.049 \quad V = 0.25 \quad \alpha = 0.052 \quad (17)$$

$$V = 0.2 \quad \alpha = 0.058 \quad V = 0.17 \quad \alpha = 0.066 \quad (18)$$

It should be noted that  $V = 0.17$  corresponds to the last limiting point to which all the values  $V > 0.17$  accumulate. This point is a focus; therefore, the “quantization” of thermodynamics by the tunnel canonical operator [11] strongly erodes this point.

The rotation is performed at points on the isotherm–isochore  $T_r = 1, P_r = 1$ . This allows us to interpret the measurements of the new variables  $\tilde{P}_r, \tilde{V}_r$  as the “new pressure” and “new volume,” because the critical vertical has not been violated. This is related to the fact that the measurement of pressure in a fluid, just as in a liquid, takes into account the actual contribution of collisions between the measuring device and the monomers as well as that of cluster oscillations. The relationship between the measured volume and the fractal dimension was discussed above.

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