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A Mathematical Theory of the Supercritical State Serving as an Effective Means of Destruction of Chemical Warfare Agents[*]

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Abstract—It is well known that the supercritical state of a gas has great dissolving capacity. In this paper, the mathematical reason for this phenomenon is studied in great detail.

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The journal *Mathematical Notes* is, of course, a purely mathematical journal. However, the author, being a mathematician, cannot turn a blind eye towards issues of vital importance if a mathematical theory leads to their solution. In this case, I am referring to the application of chemical warfare agents and the problem of their dissolution in other gases.

It is well known that dissolving capacity usually increases with temperature. The dissolving capacity of solvents increases for high pressures at which gas density is comparable with liquid density. In compressed gases, substances dissolve to a considerable degree.

But, at high pressures and high temperatures, a gas becomes a fluid, which is something in between a liquid and a gas and contains a large number of clusters. It is known that fluids are very good solvents. The question of the physical reason for this fact was discussed in the author's paper [1]. It turns out that, in a fluid, in addition to the "normal" component due to clusters, there is also a superfluid component consisting of monomers, isolated molecules, i.e., essentially of the same gas. And these monomers pass through the normal component "without friction," i.e., as a superfluid. This effect of the "two-liquid model" is not related at all to the effect of superfluidity and to the Thiess–Landau two-liquid model for liquid helium. However, in both cases, we deal with the same mathematical model, namely, a model of two almost unmixed series (an idea due to Landau, which was mathematically justified and thoroughly developed by Bogolyubov).

Many series (as an example, the famous Balmer series) appear in various physical problems. If the lowest energy level of one of the series is significantly higher than that of another series, then, mathematically, we obtain the same pattern as in the case of superfluid helium. But, physically, these can be, for example, two strings in weak resonance with each other, the first of which is in a medium where it is always excited and its lowest energy level is higher than that of the second string.

In the case of a fluid, the normal cluster component constitutes a sort of cell-like, constantly varying chaotic structure, which is penetrated, without friction, by the gas component, a solvent, at high pressure and temperature (but at which the molecules do not not break up, forming a plasma). Hence fluids contain a compressed gas component at sufficiently high temperature; it is this component that could be effectively used to destroy chemical warfare agents.

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1. NORMAL SUPERCRITICAL STATE

Following Einstein, various textbooks on physics state that, for a Bose gas, a Bose condensate appears whenever the chemical potential is zero. This effect arises in classical gas, because, as we have shown, all the mathematical relations for an ideal gas coincide with those for a quantum Bose gas[1].

As was already stated by the author in [2], for supercritical states, we obtain a new distribution, which is called a parastatistical distribution or a Gentile distribution [3].

Until the most recent experiments, the states at $P > P_c$ and $T > T_c$ were regarded as supercritical. As pointed out in [2], they contain the difference of two polylogarithms of the same rank, and, in them, the passage to the positive chemical potential is possible.

In the case of a new ideal gas and parastatistics, we assume that the density (i.e., the number of particles N) is independent of the number of clusters created as a result of the increase in the temperature T_r. The following condition is imposed on the value of $K(T_r, \gamma)$:

$$\rho_c^{(\gamma)} = \frac{N_c^{(\gamma)}}{V} = C(\gamma)\zeta(1+\gamma) = T_r^{1+\gamma}C(\gamma)\left[1-\left(\frac{1}{1+K}\right)^{\gamma}\right]\zeta(1+\gamma). \tag{1}$$

It leads to a relation for $K(T_r, \gamma)$ for $\mu = 0$ (see (2)).

In the case where K is not large, we use the dependence $K(T_r)$, which, for $\mu = 0$, is defined by the formula

$$T_r = (1-(K+1)^{-\gamma})^{-1/(\gamma+1)}, \qquad \gamma = \gamma(T_r). \tag{2}$$

under the additional condition $Z|_{\mu=0} = 1$:

$$\frac{\left\{1-\left(K(T_r)+1\right)^{-\gamma(T_r)-1}\right\}^{-1/(\gamma(T_r)+2)}}{\left\{1-\left(K(T_r)+1\right)^{-\gamma(T_r)}\right\}^{-1/(\gamma(T_r)+1)}} \cdot \frac{\zeta(2+\gamma(T_r))}{\zeta(1+\gamma(T_r))} = 1. \tag{3}$$

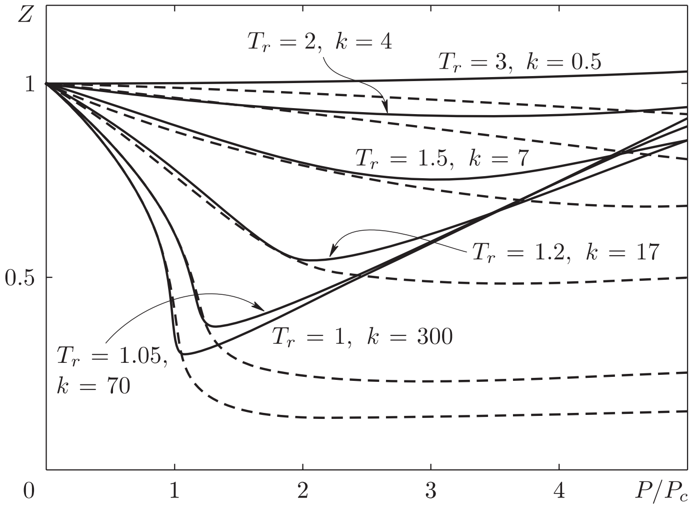

Fig. 1. Isotherms for $\gamma = 0.312$. The solid lines depict the Van-der-Waals isotherms and the dotted lines depict the parastatistical isotherms corresponding to the different values of k.

The spinodal and $K(T_r)$ are obtained from the parastatistic regarded as the critical isochore issuing from the point $Z = Z_c \approx 0.29$ for methane; see Fig. 2. The spinodal turns out to be similar to isochore 0.30 in Fig. 2 in the sense that if the maximal value of K is $N = 100$ (see Fig. 1), then the critical parastatistical spinodal will be in a sufficiently good agreement with the experimental isochore.

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[1] This was noted in a footnote in Sec. 54 "Nonequilibrium Fermi and Bose gases" in the book [9].

Figure 1, shows that the parastatistic is in good agreement with the supercritical values of the Van-der-Waals model on the first, main sheet of the Z, P-diagram. After reaching their minima, the curves corresponding the Gentile statistic differ strongly. When we pass to the second sheet of the density for $\mu = 0$, the pattern significantly changes in view of the "reflection principle." Here we do not use the special Bachinskii conditions on the Zeno line $Z = 1$, because they are obtained by taking into account the intermolecular potential [4].

It is of interest to note that, for an ideal noninteracting gas, purely statistical effects produce a similar pattern: the Boyle temperature $T_B = 2.5T_c$, the critical isochore is similar to the experimental one, etc.

Of course, it would be nice to define the percentage of the gas superfluid component. But this is impossible without the knowledge of the interaction potential [5].

Note that, for $T_r > 1.5$, the condition $P_r = 1$ for the supercritical pattern does not hold in experiments. The value of supercritical pressure increases with temperature very rapidly.

For the temperature $T_r > 1.5$, the supercritical fluid [6] even exits from the domain $Z \le 1$. Therefore, in our case, the supercritical pattern must considered only at temperatures close to the critical temperature.

In addition, note that the passage to another distribution (from a Bose–Einstein type distribution to the Gentile distribution, the parastatistic) requires, in general, a new definition of the constant of the Ω-potential. Apparently, these constants must be identical to match these two Ω-potentials. However, as shown in [2] and in [7], the remark above, as well as the principle of preference, significantly changes the way this constant depends on $\gamma(T)$.

We now pass from the integral $\int_0^\infty$ to $\int_0^A$, i.e.,

$$\begin{aligned}\int_0^\infty &\biggl(\frac{1}{e^{bx-b\mu}-1}-\frac{N}{e^{N(bx-b\mu)}-1}\biggr)x^\gamma\,dx\\ &=\int_0^A\biggl(\frac{1}{e^{bx-b\mu}-1}-\frac{N}{e^{N(bx-b\mu)}-1}\biggr)x^\gamma\,dx+\int_A^\infty\biggl(\frac{1}{e^{bx-b\mu}-1}-\frac{N}{e^{N(bx-b\mu)}-1}\biggr)x^\gamma\,dx\\ &=\int_0^A\biggl(\frac{1}{e^{bx-b\mu}-1}-\frac{N}{e^{N(bx-b\mu)}-1}\biggr)x^\gamma\,dx+O(e^{-b(A-\mu)}A^\gamma)+O(e^{-bN(A-\mu)}A^\gamma).\end{aligned}$$

Let us consider the negative values of γ.

Lemma. *Consider the integral*

$$I = B\int_0^A\biggl(\frac{1}{e^{bx-b\mu}-1}-\frac{k_0}{e^{k_0(bx-b\mu)}-1}\biggr)x^\gamma\,dx \tag{4}$$

where $-1 < \gamma < 0$, *B is a constant depending on b, and A is a constant much greater than* 1.

Then

$$I = -\frac{B}{b^{\gamma+1}}c^{(Ab)}_{b\mu,\gamma}+\frac{Bk_0^{-\gamma}}{b^{\gamma+1}}c^{(k_0Ab)}_{k_0b\mu,\gamma}, \tag{5}$$

where

$$c^{(A)}_{\mu,\gamma} = \int_0^A\biggl(\frac{1}{\xi-\mu}-\frac{1}{e^{\xi-\mu}-1}\biggr)\xi^\gamma\,d\xi. \tag{6}$$

Proof. Note that the value of B does not affect calculations in any way (it appears as a multiplier both in the original and final relations); therefore, in the proof, we set $B = 1$.

Let us make the change of variable $\xi = bx$ in the integral (4). Then

$$I = \frac{1}{b^{\gamma+1}}\int_0^{Ab}\biggl(\frac{1}{e^{\xi-b\mu}-1}-\frac{k_0}{e^{k_0(\xi-b\mu)}-1}\biggr)\xi^\gamma\,d\xi.$$

Adding and subtracting the summands, we express the integral as the sum of the following three integrals:

$$\frac{1}{b^{\gamma+1}}\int_0^{Ab}\biggl(\frac{1}{e^{\xi-b\mu}-1}-\frac{1}{\xi-b\mu}\biggr)\xi^\gamma\,d\xi+\frac{1}{b^{\gamma+1}}\int_0^{Ab}\biggl(\frac{1}{\xi-b\mu}-\frac{1}{(\xi-b\mu)(1+\frac{k_0}{2}(\xi-b\mu))}\biggr)\xi^\gamma\,d\xi$$
$$-\frac{k_0^{-\gamma}}{b^{\gamma+1}}\int_0^{Ab}\biggl(\frac{k_0^{\gamma+1}}{e^{k_0(\xi-b\mu)}-1}-\frac{k_0^{\gamma+1}}{k_0(\xi-b\mu)(1+\frac{k_0}{2}(\xi-b\mu))}\biggr)\xi^\gamma\,d\xi. \tag{7}$$

Introducing the notation (6), we find that the first integral from (7) is $-c_{b\mu,\gamma}^{(Ab)}/b^{\gamma+1}$.

In the second integral (7), we subtract the fractions and then make the change $\eta=k_0\xi$, obtaining

$$\frac{1}{b^{\gamma+1}}\int_0^{Ab}\biggl(\frac{1}{\xi-b\mu}-\frac{1}{(\xi-b\mu)(1+\frac{k_0}{2}(\xi-b\mu))}\biggr)\xi^\gamma\,d\xi=\frac{1}{b^{\gamma+1}}\int_0^{Ab}\frac{1+\frac{k_0}{2}(\xi-b\mu)-1}{(\xi-b\mu)(1+\frac{k_0}{2}(\xi-b\mu))}\xi^\gamma\,d\xi$$
$$=\frac{1}{b^{\gamma+1}}\frac{k_0}{2}\int_0^{Ab}\frac{1}{(1+\frac{k_0}{2}(\xi-b\mu))}\xi^\gamma\,d\xi=\frac{1}{b^{\gamma+1}}\frac{(k_0)^{-\gamma}}{2}\int_0^{Abk_0}\frac{1}{1+\frac{\eta-k_0b\mu}{2}}\eta^\gamma\,d\eta=\frac{k_0^{-\gamma}c_1}{b^{\gamma+1}},$$

where we have introduced the notation

$$c_1=\int_0^\infty\frac{1}{2(1+\frac{\eta-k_0b\mu}{2})}\eta^\gamma d\eta.$$

In the third integral from (7), we make the same change $\eta=k_0\xi$, obtaining

$$\frac{k_0^{-\gamma}}{b^{\gamma+1}}\int_0^{Ab}\biggl(\frac{k_0^{\gamma+1}}{e^{k_0(\xi-b\mu)}-1}-\frac{k_0^{\gamma+1}}{k_0(\xi-b\mu)(1+\frac{k_0}{2}(\xi-b\mu))}\biggr)\xi^\gamma\,d\xi$$
$$=\frac{k_0^{-\gamma}}{b^{\gamma+1}}\int_0^{Abk_0}\biggl(\frac{1}{e^{\eta-k_0b\mu}-1}-\frac{1}{(\eta-k_0b\mu)(1+\frac{\eta-k_0b\mu}{2})}\biggr)\eta^\gamma\,d\eta$$
$$=\frac{k_0^{-\gamma}}{b^{\gamma+1}}\int_0^{Abk_0}\biggl(\frac{1}{e^{\eta-k_0b\mu}-1}-\frac{1}{\eta-k_0b\mu}+\frac{1}{2(1+\frac{\eta-k_0b\mu}{2})}\biggr)\eta^\gamma\,d\eta$$
$$=\frac{k_0^{-\gamma}}{b^{\gamma+1}}\biggl(\int_0^{Abk_0}\biggl(\frac{1}{e^{\eta-k_0b\mu}-1}-\frac{1}{\eta-k_0b\mu}\biggr)\eta^\gamma\,d\eta+\int_0^{Abk_0}\frac{1}{2(1+\frac{\eta-k_0b\mu}{2})}\eta^\gamma\,d\eta\biggr)$$
$$=\frac{-k_0^{-\gamma}c_{k_0b\mu,\gamma}^{(Abk_0)}}{b^{\gamma+1}}+\frac{k_0^{-\gamma}c_1}{b^{\gamma+1}}.$$

Collecting all the expressions together, we find that the sum of the three integrals (7) is

$$-\frac{1}{b^{\gamma+1}}c_{b\mu,\gamma}^{(Ab)}+\frac{k_0^{-\gamma}}{b^{\gamma+1}}c_1+\frac{k_0^{-\gamma}}{b^{\gamma+1}}c_{k_0b\mu,\gamma}^{(Abk_0)}-\frac{k_0^{-\gamma}}{b^{\gamma+1}}c_1=-\frac{1}{b^{\gamma+1}}c_{b\mu,\gamma}^{(Ab)}+\frac{k_0^{-\gamma}}{b^{\gamma+1}}c_{k_0b\mu,\gamma}^{(Abk_0)},$$

i.e., as a result, the integral c_1 is cancelled out. ☐

It follows from the number-theoretic problem of type "partitio numerorum" considered in [11] as well as from [12]–[14] that, in the thermodynamic conception, the temperature T is a large parameter, $T\to\infty$. This fact is not always understood by physicists. In addition, the number of particles N is a also large parameter (this is well known) and the energy ε, being the sum $\varepsilon=\sum\varepsilon_iN_i$, is a large parameter, too. In number theory, all quantities are dimensionless. Obviously, $\varepsilon/\varepsilon_0\gg N$.

Consider the case $\tilde\mu=0$ and $\gamma<0$.

We introduce the following notation for the integrand in (6):

$$F(\xi)=\biggl(\frac{1}{\xi}-\frac{1}{e^\xi-1}\biggr).$$

For $\gamma < 0$ and $\tilde{\mu} = 0$, we have the "reflection principle"

$$N_c = \Lambda^{\gamma-\gamma_c}\sum_{j=1}^{\infty}\frac{j^{\gamma}}{e^{bj}-1} = \Lambda^{\gamma-\gamma_c}\biggl\{\sum_{j=1}^{\infty} j^{\gamma}\frac{1}{bj} - \sum_{j=1}^{\infty} j^{\gamma}F(bj)\biggr\}, \qquad b = \frac{1}{T}. \tag{8}$$

$$\sum_{j=1}^{\infty} j^{\gamma}F(bj) = \sum_{j=1}^{\infty} f(j) \le \int_0^{\infty} f(x)\,dx = \int_0^{\infty} x^{\gamma}F(bx)\,dx = b^{-\gamma-1}\int_0^{\infty} x^{\gamma}F(x)\,dx. \tag{9}$$

By the definition of the Riemann ζ function in terms of a series, we have

$$\sum_{j=1}^{\infty} j^{\gamma}\frac{1}{bj} \equiv T\zeta(1+|\gamma|).$$

Thus,

$$N|_{\tilde{\mu}=0} = \Lambda^{\gamma-\gamma_c}T\zeta(1-\gamma) + O(b^{-1-\gamma}), \qquad \gamma < 0, \quad b \to 0, \tag{10}$$

where ζ is the Riemann zeta function.

Relation (9) for the second sheet $\gamma < 0$ implies the following most important relation between the large parameters N and T:

$$\frac{N}{T} = \zeta(1+|\gamma|). \tag{11}$$

In calculating the "density," we essentially use this asymptotic equality on the second sheet. As was already repeatedly stated, for the density, we now pass to the second sheet on the spinodal $\mu = 0$ in the supercritical pattern (the parastatistic, or the Gentile statistic). Here, simultaneously with the change of the sign of the chemical potential at the Bose condensate point, the number of degrees of freedom experiences a jump from $D = 2\gamma + 2$ to $D_1 = 2 - 2\gamma$.

We apply the lemma on density, as well as the equality $Nb = \zeta(1-\gamma)$, obtaining

$$\begin{aligned}\rho &= B\int_0^{A}\biggl(\frac{1}{e^{bx-b\mu}-1} - \frac{N}{e^{N(bx-b\mu)}-1}\biggr)x^{\gamma}\,dx = \frac{BN^{-\gamma}}{b^{\gamma+1}}\int_0^{AbN}\biggl(\frac{1}{\xi-b\mu N} - \frac{1}{e^{\xi-b\mu N}-1}\biggr)\xi^{\gamma}\,d\xi \\ &= \frac{BN^{-\gamma}}{b^{\gamma+1}}\int_0^{A\zeta(1-\gamma)}\biggl(\frac{1}{\xi-\mu\zeta(1-\gamma)} - \frac{1}{e^{\xi-\mu\zeta(1-\gamma)}-1}\biggr)\xi^{\gamma}\,d\xi,\end{aligned} \tag{12}$$

where $\zeta(1-\gamma) = \zeta(1.222) = 5.097$.

Similarly, for $K < N$ ($K = (K/N)N$), we have

$$\begin{aligned}\rho &= B\int_0^{A}\biggl(\frac{1}{e^{bx-b\mu}-1} - \frac{K}{e^{K(bx-b\mu)}-1}\biggr)x^{\gamma}\,dx = \frac{BK^{-\gamma}}{b^{\gamma+1}}\int_0^{AbK}\biggl(\frac{1}{\xi-b\mu K} - \frac{1}{e^{\xi-b\mu K}-1}\biggr)\xi^{\gamma}\,d\xi \\ &= \frac{BK^{-\gamma}}{b^{\gamma+1}}\int_0^{A\frac{K}{N}\zeta(1-\gamma)}\biggl(\frac{1}{\xi-\mu\frac{K}{N}\zeta(1-\gamma)} - \frac{1}{e^{\xi-\mu\frac{K}{N}\zeta(1-\gamma)}-1}\biggr)\xi^{\gamma}\,d\xi.\end{aligned} \tag{13}$$

In the last integral, we take $A = N$.

Remark 1. As is seen from the lemma and the reflection principle, there is no volume on the second sheet. As was pointed out previously, volume in thermodynamics usually counterbalances the number of particles N, which is an "infinitely large" quantity. In what units must we measure the volume so as to counterbalance N? In nanometers? In angstroms?

As a matter of fact, in experiments, density is determined as $\rho = N/V$. However, on the second sheet, just as in negative pressures, holes-pores appear and density, as a quantity counterbalancing the number N, is no longer suitable: the density of pumice is not proportional to the number of particles.

As was already stated in the physical literature, the second sheet for $\gamma \le 0$ spreads onto the quadrant $-P, -Z$.

Generally speaking, there is also no volume in the potential Ω:

$$d\Omega = -S\,dT - \mu\,dN$$

(see [9, Sec. 24, formula (24.14)]).

Therefore, on the second sheet, the "infinitely large" parameter N is counterbalanced by the "infinitely large" parameter T (the temperature). The last fact is strange and can be explained, in particular, by the fact that the temperature $T = 0$ is unattainable.

As was already stated in great detail [15], it is more informative to consider $\ln T$ when $\ln 0 = -\infty$. The use of this quantity ($-\infty$) as a reference point always yields an infinitely large quantity. In other words, if we measure temperature in Kelvins, then T_c has, a rule, values of order 10^3, but if the units $1/10^{20}$ Kelvin are used, then the temperature is 10^{23}.

Only the approach mapped out above allows us to extend pressure and the compressibility factor into the negative domain.

In addition, note that the main relation of thermodynamics

$$\sum_{ij} N_{ij} = N, \qquad \sum iG_i N_i = \varepsilon, \tag{14}$$

where

$$N_i = \sum_{k=1}^{q_i} N_{ik}, \qquad q_i = \left[\frac{\Gamma(i + D/2)}{\Gamma(D/2)\Gamma(i+1)}\right],$$

also does not contain volume.

Since these equations are number-theoretic Diophantine equations, it follows that mathematicians have known for a long time how to deal with infinitely large quantities as $N \to \infty$. The main formula has been proved, but there is no "derivation" of the formula required by the physicists. The author of this formula Ramanujan stated that it was revealed to him by the goddess Radha in his sleep.

We showed in [7] that the dimension of the Bose distribution with respect to momenta coincides with the number of degrees of freedom D defined for an ideal Boltzmann–Maxwell Bose gas for which

$$c_v = \frac{D}{2}, \qquad c_p = 1 + c_v, \qquad \tilde{\gamma} = \frac{c_p}{c_v} = \frac{D+2}{D}.$$

Naturally, the degrees of freedom are related to momenta. Although no attention had been paid to this correspondence before the author's papers dealing with this subject were published, it looks quite natural.

But $p^2/(2m)$ is equal to the kinetic energy ε. Therefore, if we take the integral over the energy ε, then the dimension (the measure) over which the integral is taken, is decreased twofold. In the final analysis, such an integral is not only not less natural than an integral over momenta, but, eventually, in the book [9], the authors come to an integral over ε-energy (see the section "Bose and Fermi gases" and the beginning of Sec. 2).

In the main relation

$$\sum N_i \varepsilon_i = \varepsilon, \qquad \sum N_i = N, \tag{15}$$

and ε_i is the value of the energy.

Thus, it is more logical to talk about dimension not with respect to momenta, but with respect to energy. Then the dimension with respect to energy will be two times less than the dimension with respect to momenta, and it is

$$D_{ener} = \frac{D_{imp}}{2} = \gamma + 1.$$

From a mathematical or, more precisely, from a logical-mathematical point of view, this is more natural.

In our case, the discrete variable is energy, not momenta, and the dimension 1 corresponds to the natural series. Recall Kronecker's words: "God created integers, and everything else is Man's creation." This remark is especially significant to bear in mind when passing to negative pressures (see [10]).

First, in the Bose–Einstein distribution, these considerations allow us to replace the kinetic energy $p^2/2m$ by the general Hamiltonian, which includes, for example, potential energy. A more significant generalization given in [1] consists in the consideration of a pairwise interaction potential, and a self-consistent potential is substituted into the original formula.

In the book [9], in the final formulas, the measure under the integral over energy in the formula for the number of particles is multiplied by the energy raised to the first power (see [9, formulas (55.5), (55.7)]). In the original formula for the series from which we pass to the integral, this corresponds to multiplication by i, the eigenvalue number. This corresponds to the spectrum of an oscillator up to a multiplier. In this sense, we can say that we now pass to the oscillatory (photon) representation. Accordingly, G_i corresponds to the multiplicity (degeneracy) of the spectrum.

Since, so far, we have considered gas without interaction, it follows that the multiplicities of the eigenvalue i (in the mean) are $i^{\gamma+1}$. In the case of an external potential, we must again pass to the "oscillatory representation" and take into account the new multiplicities corresponding to the action of the external field. This will be considered in detail in the next section.

Remark 2 (On some coincidences)**.** We have already seen in Fig. 1 that the real maximal number of particles $K(1)$ is equal to 300. Thus, $N = K(1)$ is not equal to infinity, but is equal to 300. Accordingly, we must renormalize all the curves and temperatures, taking into account the fact that the maximal number of particles occupying the energy level with number i, is equal to 300. In view of the identity $N = \zeta(1 + |\gamma|)T$, this changes the values of $K(T)$. The value $\gamma = 0.222$ corresponds to Fig. 2.

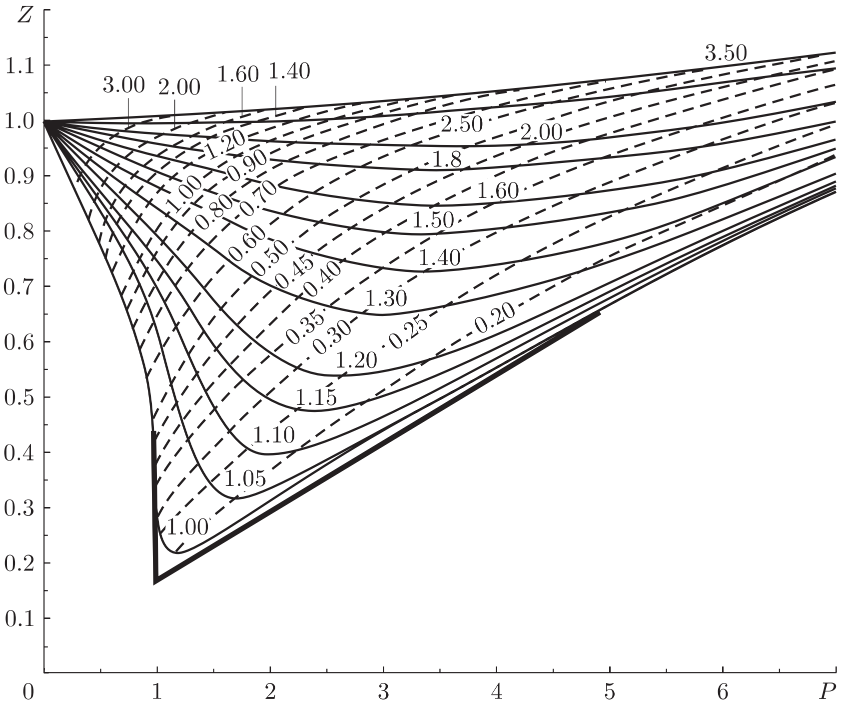

Fig. 2. Isotherms (the solid lines) and isochores (the dotted lines) for methane in the supercritical states. The bold line provides the angle of the tangent to the critical isotherm at the point $Z = 1$ with the Z axis.

For $K = 100$, the critical isotherm obtained from our formulas coincides well near the minimum with Fig. 2, while the point of intersection of the "critical" isotherm ($N = K = 100$) with the line $Z = 1$ is $P = 6$, i.e., it again turns out to be somewhat greater than that shown in Fig. 2.

Recall that we are referring to noninteracting particles, and all these "random" coincidences are due to the remarkable property of Bose condensate, a property discovered by Einstein.

2. THE SUPERFLUID SUPERCRITICAL STATE

Let the spectral measure of the self-adjoint operator dE_λ be absolutely continuous with respect to the measure $d\lambda$. We call the measure $d\lambda$ an oscillatory measure, because the line $\lambda \geq 0$ corresponds to the natural series $i = 0, 1, \dots, n, \dots$, and the natural series corresponds to the spectrum of an oscillator [16]. In this oscillatory representation, we follow the philosophic-logical ideology of Kronecker and Poincaré.

Self-consistent field (mean field) operators are defined by using the pairwise interaction operator between the particles and, after the introduction of the so-called dressed potential

$$W = u(x) + \int V(x-\xi)\rho(\xi)\,d\xi, \qquad x \in R^3, \quad \xi \in R^3, \qquad V(x) \in C^\infty, \quad u(x) \in C^\infty, \tag{16}$$

they become "almost" self-adjoint operators of Schrödinger type, where $\rho(*) = \Psi^*(x)\Psi(x)$ is the density of the distribution. The author has called these operator unitarily nonlinear [17].

As was already stated in [1], the series of such an operator are determined, as is customary, by using the variational equations. We consider classical gas and, therefore, the small parameter $\hbar$, the Planck constant, must tend to zero. However, as has already been pointed out, it is multiplied by a parameter that can be arbitrarily large and, therefore, in this rather rare case, it remains in the classical limit. It is this term that gives the superfluid series.

The analog of the thermodynamic potential of a system of noninteracting particles for the discrete variant is of the form

$$\Omega = \frac{-T^{\gamma+2}}{\Lambda^{(1+\gamma)}} \sum_i \ln \sum_{n=0}^{N} g_i \biggl(\exp - \Bigl(\varkappa + \frac{h\pi\omega i}{T_r}\Bigr)\biggr)^n, \tag{17}$$

where T_r is the temperature, μ is the chemical potential, $\varkappa = \mu/T$, Λ is a dimensionless constant, g_i is the statistical weight of the ith state: $g_i = i^{D/2}$, and D is the fractional dimension of the Bose–Einstein type distribution and ω is the frequency of the oscillator.

If we consider two variables of the natural series i and j, then the terms of the sum

$$\sum_{ij} N_{ij} = N$$

can be regrouped as follows: for two summands, $i + j = l$, for three summands, $i + j + k = l$, etc.

Then $\sum_{ij} N_{ij}$ can be expressed as $\sum G_l N_l$, where G_l is the statistical weight corresponding to the natural series N_l. In eigenvalue problems, this is called the multiplicity of the spectrum. In our example, the spectrum proportional to the natural series is the spectrum of the operator corresponding to an oscillator, and the statistical weights will correspond to the multiplicities (degeneracy) of the spectrum.

The difference of a statistical weight from multiplicity is that the statistical weight can be noninteger. For example, if the multiplicity oscillates depending on l, then the statistical weight is the mean over these oscillations. This can be regarded as the fractional dimension of the space D.

Let us write the thermodynamic potential Ω[2] of Bose gas of fractional dimension $D = 2(1+\gamma)$ as

$$\Omega(\mu, T) = \frac{T^{2+\gamma}}{\Lambda^{1+\gamma}\Gamma(2+\gamma)} \int_0^\infty \frac{t^{1+\gamma}\,dt}{(e^t/a) - 1} = \frac{-T^{2+\gamma}}{\Lambda^{1+\gamma}} \operatorname{Li}_{2+\gamma}(a), \tag{18}$$

where T is the temperature, $a = \exp(\mu/T)$ is the activity, μ is the chemical potential, Γ is the gamma function, and Λ is a constant to be define later.

The function $\operatorname{Li}_s(a)$ introduced in (18) is called a polylogarithm and defined by

$$\operatorname{Li}_s(x) = \frac{1}{\Gamma(s)} \int_0^\infty \frac{t^{s-1}}{(e^t/x) - 1}, \qquad \operatorname{Li}_s(1) = \zeta(s), \tag{19}$$

where $\zeta(s)$ is the Riemann zeta function.

The total number of trials N is

$$N = -\frac{\partial\Omega}{\partial\mu} = \frac{T^{1+\gamma}}{\Lambda^{(1+\gamma)}} \operatorname{Li}_{1+\gamma}(a).$$

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[2] We preserve the thermodynamic notation for the Ω-potential used in [9, Sec. 24]. The Ω-potential is the action $\mathbb{S}$ on the 2-dimensional Lagrangian manifold in 4-dimensional $(T, S; N, \mu)$ phase space. On the other hand, we have $d\Omega = -S\,dT - \mu\,dN$.

Since, in view of the relation

$$\sum_{ij} N_{ij} = \sum_{j} G_j N_j = N, \tag{20}$$

we have $N_{ij} \le N$, it follows that this condition is not an additional constraint. Summing the finite geometric progression, we obtain

$$\Omega_i(k) = \frac{-T}{\Lambda^{(1+\gamma)}} \ln \sum_{n=0}^{N} g_i \biggl(\exp\Bigl(-\varkappa - \frac{h\pi\omega i}{T_r}\Bigr)\biggr)^n = \frac{T}{\Lambda^{(1+\gamma)}} \ln g_i \frac{1-\exp(-\varkappa - \frac{h\pi\omega i}{T_r})(N+1)}{1-\exp(-\varkappa - \frac{h\pi\omega i}{T_r})}, \tag{21}$$

where $g_i = i^{\gamma+1}$ and $\varkappa = \mu/T$.

The potential Ω is equal to the sum of Ω_i over i:

$$\Omega = \sum \Omega_i, \quad d\Omega = -S\,dT - N\,d\mu, \tag{22}$$

where S is the entropy.

For the total number of trials, we have the formula $N = -\partial\Omega/\partial\mu$. As a result, for $\gamma \le 0$ we obtain[3]

$$N = \frac{1}{\Lambda^{\gamma-\gamma_c}} \sum_i \biggl(\frac{i^\gamma}{\exp\,(\varkappa + \frac{h\pi\omega i}{T_r}) - 1} - \frac{(N+1)i^\gamma}{\exp\,[(N+1)(\varkappa + \frac{h\pi\omega i}{T_r})] - 1}\biggr), \tag{23}$$

where Λ is the constant, $\varkappa = -\tilde{\mu}/T$, and $\tilde{\mu}$ is the chemical potential of the Bose-condensate state, i.e., for $-1 < \gamma \le 0$.

Theorem. *Suppose that $\alpha = \gamma + 1$, $k = N + 1$, and $b = 1/T_r$. The following relation holds*:

$$\sum_j \Bigl(\frac{j^\gamma}{e^{bj+\varkappa} - 1} - \frac{kj^\gamma}{e^{bkj+k\varkappa}}\Bigr) = \frac{1}{\alpha}\int_0^\infty \Bigl(\frac{1}{e^{bx+\varkappa} - 1} - \frac{k}{e^{bkx+k\varkappa} - 1}\Bigr)\,dx^\alpha + R,$$

where $R = O(b^{-\alpha})$.

Proof. By the Euler–Maclaurin formula, we have

$$|R| \le \frac{1}{\alpha}\int_0^\infty |f'(x)|\,dx^\alpha, \qquad \text{where} \quad f(x) = \frac{1}{e^{bx+\varkappa} - 1} - \frac{k}{e^{k(bx+\varkappa)} - 1}.$$

Let us calculate the derivative, obtaining

$$\begin{gathered} f'(x) = \frac{bk^2 e^{k(bx+\varkappa)}}{(e^{k(bx+\varkappa)} - 1)^2} - \frac{be^{bx+\varkappa}}{(e^{bx+\varkappa} - 1)^2}, \\ |R| \le \frac{1}{\alpha b^\alpha}\int_0^\infty \Bigl|\frac{k^2 e^{k(y+\varkappa)}}{(e^{k(y+\varkappa)} - 1)^2} - \frac{e^{y+\varkappa}}{(e^{y+\varkappa} - 1)^2}\Bigr|\,dy^\alpha. \end{gathered} \tag{24}$$

We also have

$$\frac{e^z}{(e^z - 1)^2} = \frac{1}{z^2} + \psi(z),$$

where $\psi(z)$ is a smooth function and $|\psi(z)| \le C(1 + |z|)^{-2}$. Setting $z = y$ and $z = ky$, we obtain the following estimate:

$$\begin{aligned} |R| &\le \frac{1}{\alpha b^\alpha}\int_0^\infty \bigl|\psi\bigl(k(y+\varkappa)\bigr) - \psi(y+\varkappa)\bigr|\,dy^\alpha \\ &\le \frac{k^{-\alpha}}{b^\alpha}\int_{k\varkappa}^\infty |\psi(y)|\,dy^\alpha + \frac{1}{b^\alpha}\int_\varkappa^\infty |\psi(y)|\,dy \le Cb^{-\alpha} \end{aligned} \tag{25}$$

with some constant C.

The theorem is proved. □

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[3] In rigorous terms, the fact that the power of g_i decreases strictly by 1 follows from the fact that the "energy" levels ε_i in our case are i and that, after taking the derivative with respect to μ, the power of T decreases by 1.

Corollary 1. *Taking into account the fact that the term containing* $N+1$ *in* (21) *tends to zero as* $N \to \infty$*, we obtain the following relation*:

$$M = \frac{\Lambda^{\gamma_c-\gamma}}{\alpha\Gamma(\gamma+2)} \int \frac{\xi\, d\xi^\alpha}{e^{b\xi}-1} = \frac{\Lambda^{\gamma_c-\gamma}}{b^{1+\alpha}} \int_0^\infty \frac{\eta d\eta^\alpha}{e^\eta-1}, \tag{26}$$

where $\alpha=\gamma+1$, $b=1/T_r$, *and* $M=\varepsilon/\varepsilon_0$. *Therefore,*

$$b = \frac{1}{M^{1/(1+\alpha)}} \bigg(\frac{\Lambda^{\gamma_c-\gamma}}{\alpha\Gamma(\gamma+2)} \int_0^\infty \frac{\xi\, d\xi^\alpha}{e^\xi-1}\bigg)^{1/(1+\alpha)}.$$

We also have (see[18])

$$\begin{aligned}\sum_j \Big(\frac{j^\gamma}{e^{bj+\varkappa}-1} - \frac{kj^\gamma}{e^{bkj+k\varkappa}}\Big) &= \frac{1}{\alpha}\int_0^\infty \bigg\{\frac{1}{e^{b\xi}-1} - \frac{k}{e^{kb\xi}-1}\bigg\}\, d\xi^\alpha + O(b^{-\alpha}) \\ &= \frac{1}{\alpha b^\alpha}\int_0^\infty \Big(\frac{1}{e^\xi-1} - \frac{1}{\xi}\Big)\, d\xi^\alpha + \frac{1}{\alpha b^\alpha}\int_0^\infty \bigg(\frac{1}{\xi} - \frac{1}{\xi(1+(k/2)\xi)}\bigg)\, d\xi^\alpha \\ &\quad - \frac{k^{1-\alpha}}{\alpha b^\alpha}\int_0^\infty \bigg\{\frac{k^\alpha}{e^{k\xi}-1} - \frac{k^\alpha}{k\xi(1+(k/2)\xi)}\bigg\}\, d\xi^\alpha + O(b^{-\alpha}) \\ &= \frac{c(\gamma)}{b^\alpha}(k^{1-\alpha}-1) + O(b^{-\alpha}).\end{aligned}$$

Setting $k = N|_{\tilde\mu/T=0} \gg 1$, *we finally obtain*

$$N_c = N|_{\tilde\mu/T=0} \cong (\Lambda^{\gamma_c-\gamma} c(\gamma))^{1/(1+\gamma)} T_r, \tag{27}$$

where

$$c(\gamma) = \int_0^\infty \Big(\frac{1}{\xi} - \frac{1}{e^\xi-1}\Big)\xi^\gamma\, d\xi.$$

As was already stated in [1], all the arguments for the general Hamiltonian hold, in particular, for

$$\frac{p^2}{2m} + u(x), \qquad x \in R^3.$$

We can also consider the Hamiltonian operator in the semiclassical approximation.

We can also generalize the problem to the case of a self-consistent Hamiltonian, i.e., when

$$u = u\bigg(x, \int V(x-\xi)\rho(\xi)\, d\xi\bigg), \tag{28}$$

where $V(x-\xi)$ is the Lennard-Jones pairwise interaction potential (see [8]) and $\rho(\xi)$ is the density in the Vlasov equation.

The semiclassical asymptotics as $h \to 0$ of such a quantum problem no longer coincides with the original classical self-consistent Vlasov-type equation; using the complex germ method, we find that the asymptotics in question contains a semiclassical correction, which was first discovered in a paper of Bogolyubov on weakly nonideal Bose gas and which is impossible to neglect as $h \to 0$.

Now let us dwell on the notion called "collective oscillations" in classical physics and "quasiparticles" in quantum physics. This is given by the Vlasov equation for the self-consistent (or mean) field in classical physics, while, in quantum physics, by the Hartree equation (or the Hartree–Fock equation).

1) Variational equations depend on the solutions (of the original equation) near which the variations are considered. For example, in [19]–[22], we consider variations near the microcanonical distribution in the ergodic construction and, in [23]–[26], those near the nanocanonical distribution concentrated on an invariant manifold of smaller dimension, i.e., not on the manifold of constant energy, but, for example, on the Lagrangian manifold of dimension coinciding with that of the configuration space.

2) Note the following important point: the solutions of the variational equation for the Vlasov equation *do not coincide* with the classical limit for variational equations for mean-field equations in quantum theory.

Consider the quantum mean-field equation of the form

$$ih\frac{\partial}{\partial t}\varphi^t(x) = \Big(-\frac{h^2}{2m}\Delta + W_t(x)\Big)\varphi^t(x), \qquad W_t(x) = u(x) + \int V(x,y)|\varphi^t(y)|^2\,dy, \tag{29}$$

under the initial condition $\varphi|_{t=0} = \varphi_0$, where φ_0 satisfies

$$\varphi_0 \in W_2^\infty(\mathbf{R}^3), \qquad \int dx\,|\varphi_0(x)|^2 = 1.$$

To obtain the asymptotics of complex germ type [27], we must consider the system consisting of the Hartree equation (29) and the equation conjugate to it. Further, we must consider the variational system for it and, finally, replace the variations $\delta\varphi$ and $\delta\varphi^*$ by independent functions F and G. For the functions F and G, we obtain the following system of equations:

$$\begin{aligned} i\frac{\partial F^t(x)}{\partial t} &= \int dy\Big(\frac{\delta^2 H}{\delta\varphi^*(x)\delta\varphi(y)}F^t(y) + \frac{\delta^2 H}{\delta\varphi^*(x)\delta^*\varphi(y)}G^t(y)\Big), \\ -i\frac{\partial G^t(x)}{\partial t} &= \int dy\Big(\frac{\delta^2 H}{\delta\varphi(x)\delta\varphi(y)}F^t(y) + \frac{\delta^2 H}{\delta\varphi(x)\delta^*\varphi(y)}G^t(y)\Big). \end{aligned} \tag{30}$$

Note that, roughly speaking, classical equations can be derived from the corresponding quantum equations by substitutions of the form

$$\varphi = \chi e^{\frac{i}{h}S}\ \text{(WKB method)}, \qquad \varphi^* = \chi^* e^{\frac{i}{h}S^*},$$

where

$$S = S^*, \qquad \chi = \chi(x,t) \in C^\infty, \qquad S = S(x,t) \in C^\infty.$$

For a variational equation, it is natural to vary not only the limit equation for χ and χ^*, but also the functions S and S^*. This gives a new important term in the solution of the equation for collective oscillations.

Let us illustrate this fact by using the simple example studied in the famous paper of Bogolyubov dealing with "weakly nonideal Bose gas" [8].

Suppose that $u = 0$ in Eq. (29) in a 3-dimensional box with edge length L (i.e., on the torus with generatrices L, L, L); here the periodicity condition is imposed on the wave functions. Then the function

$$\varphi(x) = L^{-3/2}e^{i/h(px-\Omega t)}, \tag{31}$$

where $p = 2\pi n/L$ and n is an integer-valued vector, satisfies Eq. (29) for

$$\Omega = \frac{p^2}{2m} + L^{-3}\int dx\,V(x). \tag{32}$$

Consider the functions $F^{(\lambda)}(x)$ and $G^{(\lambda)}(x)$, where $\lambda = 2\pi n/L, n \neq 0$ and n is an integer-valued vector, of the following form:

$$\begin{aligned} F^{(\lambda)t}(x) &= L^{-3/2}\rho_\lambda e^{\frac{i}{h}|(p+\lambda)x+(\beta-\Omega)t|}, \\ G^{(\lambda)t}(x) &= L^{-3/2}\sigma_\lambda e^{\frac{i}{h}|(-p+\lambda)x+(\beta+\Omega)t|}, \end{aligned} \tag{33}$$

where

$$\begin{aligned} -\beta_\lambda\rho_\lambda &= \Big(\frac{(p+\lambda)^2}{2m} - \frac{p^2}{2m} + \widetilde{V}_\lambda\Big)\rho_\lambda + V_\lambda\sigma_\lambda, \\ \beta_\lambda\rho_\lambda &= \Big(\frac{(p-\lambda)^2}{2m} - \frac{p^2}{2m} + \widetilde{V}_\lambda\Big)\sigma_\lambda + V_\lambda\rho_\lambda, \end{aligned} \tag{34}$$

$$|\sigma_\lambda|^2 - |\rho_\lambda|^2 = 1, \qquad \widetilde{V}_\lambda = L^{-3}\int dx\, V(x) e^{\frac{i}{h}\lambda x}.$$

Using system (34), we obtain

$$\beta_\lambda = -p\lambda + \sqrt{\left(\frac{\lambda^2}{2m} + \widetilde{V}_\lambda\right)^2 - \widetilde{V}_\lambda^2}. \tag{35}$$

In this example,

$$u = e^{\frac{i}{h}s(x,t)}, \quad u^* = e^{-\frac{s(x,t)}{h}}, \qquad \text{where} \quad s(x,t,) = px + \beta t,$$

and the variation of the action for the vector $(\delta u, \delta u^*)$ is $\lambda x \pm \Omega t$.

In the more rigorous passage to the limit, we have

$$\widetilde{V}_\lambda \to V_0 = L^{-3}\int dx\, V(x).$$

Thus, in the classical limit, we obtain the famous Bogolyubov relation (35). In the case under study, $u(x) = 0$ and, just as in the linear Schrödinger equation, the exact solution coincides with the semiclassical one. The paper [26] studies the case $u(x) \neq 0$, and it turns out that a relation similar to (35) is the classical limit as $h \to 0$ for the variational equations in this general case. The curve describing the dependence of β_λ on λ is called the Landau curve; it defines the superfluid state. The value of λ_{cr} at which superfluidity disappears is called the Landau criterion.

The spectrum defined for $\lambda < \lambda_{\text{cr}}$ has the corresponding positive spectrum of the variational equation. This means that it is metastable (see [28]).

3. THE NANOCANONICAL DISTRIBUTION AND VARIATIONAL EQUATIONS

Consider the classical equation for a self-consistent field (the Vlasov equation) for the distribution function $\rho(x,p,t)$:

$$\frac{\partial\rho}{\partial t} + p\frac{\partial\rho}{\partial x} - \frac{\partial u}{\partial x}\frac{\partial\rho}{\partial p} - \alpha\frac{\partial\rho}{\partial p}\frac{\partial}{\partial x}\int V(x-\xi)\rho(\xi,\eta,t)\,d\xi\,d\eta = 0, \tag{36}$$

where $u(x) \in C^2$ and $V(x) \in C^2$ are the external potential and the interaction potential, respectively. Here

$$p\frac{\partial\rho}{\partial x} = \left(p, \frac{\partial\rho}{\partial x}\right) \equiv \sum_{i=1}^{3} p_i\frac{\partial\rho}{\partial x_i}, \qquad \frac{\partial u}{\partial x}\frac{\partial\rho}{\partial p} = \left(\frac{\partial u}{\partial x}, \frac{\partial\rho}{\partial p}\right), \qquad \text{etc.}$$

For brevity, we do not usually indicate the inner product of vectors explicitly unless such an indication is essential.

The variational equations are of the form

$$\begin{aligned}\frac{\partial\sigma}{\partial t} + p\frac{\partial\sigma}{\partial x} - \frac{\partial u}{\partial x}\frac{\partial\sigma}{\partial p} - \alpha\frac{\partial\rho}{\partial p}\frac{\partial}{\partial x}\int V(x-\xi)\sigma(\xi,\eta,t)\,d\xi\,d\eta \\ - \alpha\frac{\partial\sigma}{\partial p}\frac{\partial}{\partial x}\int V(x-\xi)\rho(\xi,\eta,t)\,d\xi\,d\eta = 0.\end{aligned} \tag{37}$$

Suppose that $\Lambda^3 = \{x = X(\alpha),\, p = P(\alpha)\}$ is the Lagrangian manifold invariant with respect to the Hamiltonian system

$$\begin{cases} \dot{x} = p, \\ \dot{p} = -\dfrac{\partial u}{\partial x} - \dfrac{\partial}{\partial x}\displaystyle\int V(x - X(\alpha))\, d\mu_\alpha, \end{cases} \tag{38}$$

(see [29], [30]) such that the corresponding Hamiltonian H is invariant on Λ^3,

$$\frac{P^2(\alpha)}{2} + u(X(\alpha)) + \int V(X(\alpha) - X(\alpha'))\, d\mu_{\alpha'} = \varepsilon = \text{const}, \tag{39}$$

where $\alpha \in \Lambda^3$ and $d\mu_\alpha$ is an invariant measure on Λ^3 [23], [24], [31].

It is well known that the δ-function on a manifold of constant energy is called a microcanonical distribution.

Suppose that δ_Λ is the δ-function on a manifold Λ. We call this distribution a nanocanonical distribution. As proved in [23], the function δ_Λ satisfies the stationary Vlasov equation (36) in the local chart on Λ. δ_Λ is of the form

$$\delta_\Lambda = \frac{\delta(p - \nabla S)}{J},$$

where

$$S(x) = \int_{\alpha_0}^{\alpha(x)} p\, dx$$

is the action on Λ^3, and J is the Jacobian $|dX/d\mu_\alpha|$.

In this case, the solution of Eq. (37) can be obtained in the form

$$\sigma = f(x)\frac{\delta(p - \nabla S)}{J} + \mathbf{a}(x)\frac{\partial}{\partial p}\frac{\delta(p - \nabla S)}{J}. \tag{40}$$

The equations for f and $\mathbf{a}$ are as follows:

$$\frac{\partial f}{\partial t} + (\nabla S, \nabla f) - (\nabla, \mathbf{a}) + (\mathbf{a}, \nabla)\ln J = 0, \tag{41}$$

$$\frac{\partial \mathbf{a}}{\partial t} + (\nabla S, \nabla)\mathbf{a} + (\mathbf{a}, \nabla)\nabla S + \nabla \int V(x - X(\alpha)) f(X(\alpha))\, d\mu_\alpha = 0. \tag{42}$$

Here we introduce the quantum correction for the equation obtained in [25],

$$\begin{aligned} \frac{\partial f}{\partial t} + (\nabla S, \nabla f) - (\nabla, \mathbf{a}) + (\mathbf{a}, \nabla)\ln J - \frac{ih}{2} J\nabla\biggl(\frac{1}{J}\nabla f\biggr) &= 0, \\ \frac{\partial \mathbf{a}}{\partial t} + (\nabla S, \nabla)\mathbf{a} + (\mathbf{a}, \nabla)\nabla S + \frac{ih}{2}\nabla\bigl\{J\bigl(\nabla, \frac{\mathbf{a}}{J}\bigr)\bigr\} + \nabla \int V(x - X(\alpha)) f(X(\alpha))\, d\mu_\alpha &= 0. \end{aligned} \tag{43}$$

In the leading term of the semiclassical asymptotics

$$f = f_0 e^{i/hS_1}, \qquad \mathbf{a} = \mathbf{a_0}\mathbf{e}^{\mathbf{i/hS_1}}, \tag{44}$$

we obtain classical variational equations. Thus, the semiclassical limit of variational equations for Hartree equations contains a superfluous term as compared to the variational equations for Vlasov equations. This superfluous term brings about the superfluidity phenomenon.

Consider the case in which $u(x) = 0$. Then the invariant Lagrangian manifold coincides with the plane $p = p_0$, while the δ-function on this manifold is of the form $\delta_\Lambda = \delta(p - p_0)$. It is easy to see that the solution of Eqs. (41)

$$f(x,t) = f_0 e^{\frac{i(\omega t + kx)}{h}}, \quad \mathbf{a}(x,t) = \mathbf{a}_0 e^{\frac{i(\omega t + kx)}{h}} \tag{45}$$

yields the famous Bogolyubov dispersion relation for Bose gas, which, in particular, describes the superfluid state of liquid helium. Thus, we have obtained the superfluid series [1], just as Bogolyubov, but only in an external field. In the classical limit, we have the term that, under the sign of the root in Bogolyubov's paper, is of the form h^2k^4 and, although $h \to 0$, the value of k can be arbitrarily large.

As pointed out at the beginning the paper, the physical meaning of this superfluid series is totally different from that in the case of helium-4.

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