

# NONEQUILIBRIUM STATISTICAL OPERATOR AS A GENERALIZATION OF GIBBS DISTRIBUTION FOR NONEQUILIBRIUM CASE

D. N. ZUBAREV

*Steklov Mathematical Institute  
Russian Academy of Sciences  
Moscow, Russia*

The history of nonequilibrium statistical operator method is presented.

The history of appearance of new ideas is a complex and an instructive process. Ideas of the theory of nonequilibrium processes are original and complicated for comprehension. Difficulties of the theory of nonequilibrium processes were formulated very clearly by R. Peierls in his lecture "Some simple notes on the basis of the transport theory" [1]. "In each theoretical analysis of transport problems it is important to imagine clearly the place at which irreversibility was carried in. If the latter isn't carried in the investigation is mistaken. Description in which the reversibility in time remains can yield both the zeroth and infinite results for conductivity, If we don't see clearly where the irreversibility is carried in, then we don't understand what we are doing."

In this lecture I will not make attempts to present the history of development of the theory of nonequilibrium processes starting from pioneer papers by Gibbs. One can find it in many books and reviews including my own book "Nonequilibrium statistical thermodynamics" [2]. I want to tell only about my way to understanding nonequilibrium statistical mechanics that led me to construction of nonequilibrium statistical operator (or functional).

For a long time I believed together with many physicists-theoreticians that general theory of nonequilibrium processes, based on the sole method like Gibbs equilibrium statistical mechanics [3], doesn't exist now and will not appear in the future and each problem needs its own method, for example, that of solving the kinetic equation (a consequent theory of kinetic equations on the basis of Gibbs method was worked out by Bogolyubov in 1946 [4]). The kinetic equation is unapplicable to liquids and therefore there is no realistic theory of fluids. These ideas are clearly expressed in the book by L. Hurevich "The principles of physical kinetics" [5] and in the course of statistical physics by L. Landau and E. Lifshitz [6].

After papers by H. Green (1952-54) [7] on the statistical mechanics of linear dissipation processes and by R. Kubo (1957) [8] on the theory of linear response of statistical systems to external perturbations, in which excellent Green-Kubo<sup>1</sup> formulae were obtained for kinetic coefficients in terms

<sup>1</sup>In 1961 these formulas were known as "Kubo formulas." At present the more exact title was accepted "Green-Kubo formulas." The theory by Green M. was further developed in papers by Mori 1958-1959 [9], who proceeded from a local equilibrium distribution on the basis of Gibbs grand canonical distribution that is more convenient to use.

of space-time correlation functions of flux densities of quantity of particles, heat and momentum, a hope arised the united general method existed. These formulae are a brilliant achievement of the theory of nonequilibrium processes. Existence of such relations lead to the natural idea, that Green-Cubo formulae are only a part of expansion of some nonequilibrium distribution function or statistical operator and they correspond to the nonequilibrium statistical ensemble. The ideas appeared in my mind in the spring of 1961 in holiday home "Solotcha" on Oka-river when I tried to generalize a review on the theory of two-time Green functions, published by me in 1960 in "Uspekhi Fizicheskikh Nauk" [10]. In that time I wasn't able to include in the review the theory of linear response of statistical systems to "thermal perturbations" in convincing form. It is impossible to represent such perturbations as a result of external field action. It seemed to me the problem of generalization of Green-Cubo formulae was interesting to a great extent but rather hopeless one, since, on the contrary to the equilibrium case, I have not had clear physical principle for construction of the nonequilibrium statistical ensemble and at the same time the inductive way using high approximations for Green-Cubo formulae seemed to be very complicated. The main idea of solving the problem came into being quite accidentally: powerful spring flood on Oka-river isolated our holiday home from railway Ryazan-Moscow on which briges were destroyed and the only exite way out appeared to be an indirect narrow-gauge line through Spas-Klepiki. The train left in the evening and I had to spend a night in a small car of narrow-gauge line so that I could sleepless not even lie down on a bench. That night I got desire to think about realization of the idea in which I trusted weakly. But it was that moment when the idea of generalization of Gibbs distribution for nonequilibrium case with the help of quasi-integrals of motion was originated.

I am going to present this idea in the form in which it arose. Quasi-equilibrium statistical operator for a stationary state of spatially inhomogeneous system has a form

$$\varrho_q = Q_q^{-1} \exp \left\{ - \int \beta(x) (H(x) - \mu(x)n(x)) dx \right\}, \quad (1)$$

where  $\beta(x) = \frac{1}{T(x)}$ ,  $T(x)$  is a temperature,  $\mu(x)$  is a chemical potential,  $H(x)$  is the operator of energy density,  $n(x)$  is the operator of particle density.

At constant temperature and chemical potential (1) transforms to the statistical operator for the grand canonical ensemble of Gibbs:

$$\varrho_0 = Q_0^{-1} \exp \left\{ - \beta(H - \mu N) \right\}, \quad (2)$$

where  $H = \int H(x)dx$ ,  $N = \int n(x)dx$  are the integrals of motion. On the other hand,  $H(x)$  and  $n(x)$  are not the integrals of motion, since they don't commute with  $H$ ,  $[H(x), H] \neq 0$ ,  $[n(x), H] \neq 0$ . The question appears if it is possible to find such operators  $A_1(x)$  and  $A_2(x)$  corresponding to  $H(x)$  and  $n(x)$ , that they should commute with  $H$ :  $[A_1(x), H] = 0$ ,  $[A_2(x), H] = 0$  and that statistical operator

$$\varrho = Q^{-1} \exp \left\{ - \int \beta(x) (A_1(x) - \mu(x)A_2(x)) dx \right\} \quad (3)$$

should commute with  $H$  and could describe correctly stationary nonequilibrium processes.

Operators  $H(x)$  and  $n(x)$  written in the Heisenberg representation

$$H(x; t) = e^{-\frac{Ht}{\hbar}} H(x) e^{\frac{Ht}{\hbar}}, \quad n(x; t) = e^{-\frac{Ht}{\hbar}} n(x) e^{\frac{HT}{\hbar}}$$

satisfy equations of motion

$$\begin{aligned} \frac{\partial}{\partial t} H(x; t) &= \frac{1}{i\hbar} [H(x; t), H] = -\text{div} \mathbf{j}_H(x; t), \\ \frac{\partial}{\partial t} n(x; t) &= \frac{1}{i\hbar} [n(x; t), H] = -\text{div} \mathbf{j}(x; t), \end{aligned} \quad (4)$$

where  $\mathbf{j}_H(x; t)$  is the density of energy flux and  $\mathbf{j}(x; t)$  is the density of particle flux in the Heisenberg representation

Natural idea appears that for construction of integrals of motion one has to add to  $H(x)$  and  $n(x)$  certain terms related to fluxes of energy and substance. It is rather easy to guess that one can try to choose as these quantities

$$\begin{aligned} A_1(x) &= H(x) - \int_0^{\infty} \text{div} \mathbf{j}_H(x; t) dt, \\ A_2(x) &= n(x) - \int_0^{\infty} \text{div} \mathbf{j}(x; t) dt. \end{aligned} \quad (5)$$

Really in the Heisenberg representation

$$\begin{aligned} A_1(x; t) &= H(x; t) - \int_0^{\infty} \text{div} \mathbf{j}_H(x; t + t_1) dt_1 = \\ &= H(x; t) - \int_t^{\infty} \text{div} \mathbf{j}_H(x; t') dt', \\ A_2(x; t) &= n(x; t) - \int_0^{\infty} \text{div} \mathbf{j}(x; t + t_1) dt_1 = \\ &= n(x; t) - \int_t^{\infty} \text{div} \mathbf{j}(x; t') dt. \end{aligned} \quad (6)$$

It follows from these relations

$$\frac{\partial}{\partial t} A_1(x; t) = 0, \quad \frac{\partial}{\partial t} A_2(x; t) = 0 \quad (7)$$

i.e.  $A_1(x) = A_1(x, 0)$  and  $A_2(x) = A_2(x, 0)$  commute one with another.

One can try to postulate the nonequilibrium statistical operator in the form

$$\varrho = Q^{-1} \exp \left\{ - \int \beta(x) (H(x) - \mu(x)n(x)) dx + \int_0^{\infty} dt \int \beta(x) (\operatorname{div} \mathbf{j}_H(x; t) - \mu(x)(\operatorname{div} \mathbf{j}(x; t))) dx \right\}. \quad (8)$$

After integration by part in the second integral of the exponent and omission of surface integrals as boundary condition we obtain the final expression for the statistical operator

$$\varrho = Q^{-1} \exp \left\{ - \int \beta(x) (H(x) - \mu(x)n(x)) dx + \int_0^{\infty} \int \mathbf{j}_H(x; t) \cdot \nabla \beta(x) - \mathbf{j}(x; t) \cdot \nabla \beta(x) \mu(x) dt dx \right\}. \quad (9)$$

These greatest notes I made in a notebook during the sleepless night (I have saved the notebook). It remained only to calculate kinetic coefficients and verify their accordance with Green-Cubo formulae at small gradients  $\beta(x)$  and  $\mu(x)$ .

After returning to Moscow I checked notes and got from (9) the linear approximation for  $\varrho$  at small  $\nabla \beta(x)$  and  $\nabla \mu(x)$  and wrote down the corresponding entropy production. But unfortunately I obtained not positive but negative entropy production. In a stationary nonequilibrium state it has to be positive and total entropy should increase since fluxes through the surface, the latter is assumed to be removed to infinity, have been omitted. I didn't find mistakes in my notes so it remained to suppose the only thing that another solution should exist except the obtained one. Let's notice that analogical situation is known in the radiation theory where two solutions of Maxwell equation exist: retarding solution and leading one. To explain the possibility of radiation one has to select the only retarding solutions by the mean of Sommerfeld's boundary condition for radiation which implies the absence of waves coming from infinity.

It was really not difficult to guess that another solution exists:

$$A_1(x) = H(x) + \int_{-\infty}^0 \operatorname{div} \mathbf{j}_H(x; t) dt, \quad (10)$$

$$A_2(x) = n(x) + \int_{-\infty}^0 \operatorname{div} \mathbf{j}(x; t) dt$$

and substituting this solution in the formula (3) we obtain the

nonequilibrium statistical operator:

$$\rho = Q^{-1} \exp \left\{ - \int \beta(x) (H(x) - \mu(x)n(x)) dx - \int_{-\infty}^0 \int (\mathbf{j}_H(x;t) \cdot \nabla \beta(x) - \mathbf{j}(x;t) \cdot \nabla \beta(x) \mu(x)) dt dx \right\}, \quad (11)$$

which leads to correct Green-Cubo formulae and to positively defined expression for the entropy production. After that I trusted in the idea of the nonequilibrium statistical operator and started to develop it with my co-workers and pupils; I deal with it so far. Now it seems to me that it is very good I've obtained first leading solutions but not retarding ones. Otherwise an illusion could arise that it is possible to obtain irreversible equations without additional hypothesis proceeding from mechanical equations.

After returning to Moscow I wrote quickly not large paper "Statistical operator for nonequilibrium systems" [11], which was represented 11.04.1961 by academician N.N.Bogolubov, my teacher and scientific guide, for publication in Reports of the Soviet Academy of Sciences. No reference was needed for publication in Reports ..., the academician's representation was sufficient. I think it was the only way to publish the formulation of the new principle in the paper that was not substantiated in full manner. The only stationary case was considered in it and there was no concrete application. The editors of the Journal of experimental and theoretical physics would decline it undoubtedly.

I'd not say the idea of the nonequilibrium statistical operator was approved by my friends. S.V.Tyablikov named it ironically Ryazan miracle. Someone may remember that in 1961 Ryazan province overfulfilled the plan of meat production fantastically after it had bought out a lot of cattle in neighbouring provinces. The latter was kept back thoroughly. Successes of the province caused canard in press. "The great achievement" was known among reporters as Ryazan miracle.

In 1961 I had an invitation to the first summer theoretical school in Odessa, organized by theoretical department of Institute of Theoretical Problems and physics department of Odessa university. I think it was the first summer school for theoreticians. In the future similar schools were held oftener. Original investigations of authors were represented for reports but their titles were not announced before. I decided to read the report on the nonequilibrium statistical distribution of Gibbs because I was interested in that how qualified physicists-theoreticians from Landau's Moscow school and Leningrad school of Institute of Physics and Technique would appreciate my paper. When in Odessa I informed the chief of organizing committee Dr. I.M.Halatnikov, that the title of my report is "The statistical operator for nonequilibrium systems" he was astonished and asked me if I was not mistaken but I confirmed the title.

My short report as it was expected caused rather intensive discussion at which except of sceptical treatment I heard also the valuable support of Dr. L.E.Gurevich (Leningrad Institute of Physics and Technique) and Dr. E.S.Fradkin (Moscow Physical Institute of the Academy of Sciences). I knew works of Gurevich due to his book "Physical kinetics" but he doubted it was possible to generalize the Gibbs theory for nonequilibrium states. (Besides that there was a lot of interesting things for me in the book by Gurevich, in particular, the theory of stationary nonequilibrium Onsager processes which

had direct relation to my report). I was acquainted with Dr. E.S.Fradkin in connection with his works on hot plasma theory because I dealt with it earlier. Dr. Gurevich treated my report first with great interest and approval but changed his mind a day later. Dr. Fradkin expressed an idea in the discussion that in order to substantiate obtained distribution one can perhaps "introduce any  $\varepsilon$ ". His proposal appeared to be correct and the problem really was further in that how to introduce "any  $\varepsilon \rightarrow 0$ " and what it meant. I tried not to take active part in the discussion personally since I have told in my report already everything that I did then due to intuitive guess and didn't have new convincing arguments.

After Odessa school I had to think have inaccuracy and non-entire what causes persuasiveness of proposed by me method for construction of the nonequilibrium statistical operator for stationary processes.

Undoubtedly it lied in that after integration by parts in spatial integrals  $\int \beta(x) \operatorname{div} \mathbf{j}_H(x; t) dx$  and  $\int \beta(x) \mu(x) \operatorname{div} \mathbf{j}(x; t) dx$  I omitted integrals over closed surface  $\int \beta(x) \mathbf{j}_H(x; t) ds$  and  $\int \beta(x) \mu(x) \mathbf{j}(x; t) ds$ , but in final results when calculating average quantities, the volume  $V \rightarrow \infty$  at fixed  $N/V$  and the surface comprising the volume  $V$  was gone to infinity. However at a stationary process integrals over the surface are not infinitesimal at fixed  $\nabla \beta$  and  $\nabla \mu$  and they are equal to the entropy production in the system. The question arises: what sense should one attach to the omission of fluxes of energy and quantity of particles through the surface? If not to treat it as an elementary mistake then one wonders if there is a possibility to formulate this process in a more accurate way with the help of two unpermutable limits:  $V \rightarrow \infty$  at fixed  $V/N$  and the limit  $\varepsilon = \frac{1}{T} \rightarrow +0$ , considering it as the boundary condition selecting retarding solutions to the Liouville equation.

It appeared to be very simple to introduce the nonequilibrium statistical operator more rigorously with the help of introducing the infinitesimal  $\varepsilon$ . Instead of relations (10) one can suppose

$$A_1^\varepsilon(x) = H(x) + \int_{-\infty}^0 e^{\varepsilon t} \operatorname{div} \mathbf{j}_H(x; t) dt = \varepsilon \int_{-\infty}^0 e^{\varepsilon t} H(x; t) dt, \quad (12)$$

$$A_2^\varepsilon(x) = n(x) + \int_{-\infty}^0 e^{\varepsilon t} \operatorname{div} \mathbf{j}(x; t) dt = \varepsilon \int_{-\infty}^0 e^{\varepsilon t} n(x; t) dt,$$

where  $\varepsilon \rightarrow +0$  is introduced after the thermodynamical transition at calculation of averages with the operator

$$\varrho_\varepsilon = Q^{-1} \exp \left\{ - \int \beta(x) (A_1^\varepsilon(x) - \mu(x) A_2^\varepsilon(x)) dx \right\}. \quad (13)$$

The values  $A_1^\varepsilon(x)$ ,  $A_2^\varepsilon(x)$  have sense of densities of energy and quantity of particles. Really

$$\int A_1^\varepsilon(x) = \int_{-\infty}^0 \varepsilon e^{\varepsilon t} \int H(x; t) dx dt = H, \quad (14)$$

$$\int A_2^\varepsilon(x) = \int_{-\infty}^0 \varepsilon e^{\varepsilon t} \int n(x; t) dx dt = N$$

and also

$$\lim_{\varepsilon \rightarrow +0} \langle A_1^\varepsilon \rangle = \langle H(x) \rangle, \quad \lim_{\varepsilon \rightarrow +0} \langle A_2^\varepsilon \rangle = \langle n(x) \rangle, \quad (15)$$

where averaging is done with the help of statistical operator satisfying the Liouville equation. From (13) and (12) we obtain

$$\rho_\varepsilon = Q^{-1} \exp \left\{ - \int \beta(x) (H(x) - \mu(x)n(x)) dx - \int_{-\infty}^0 \int e^{\varepsilon t} \beta(x) (\operatorname{div} \mathbf{j}_H(x; t) - \mu(x) \operatorname{div} \mathbf{j}(x; t)) dt dx \right\}. \quad (16)$$

After application of the Green theorem and omission of the integral over the surface we obtain the nonequilibrium statistical operator

$$\rho = Q^{-1} \exp \left\{ - \int \beta(x) (H(x) - \mu(x)n(x)) dx + \int_{-\infty}^0 \int e^{\varepsilon t} \mathbf{j}_H(x; t) \cdot \nabla \beta(x) - \mathbf{j}(x; t) \cdot \nabla \beta(x) \mu(x) dt dx \right\}, \quad (17)$$

which differs from (11) by the factor  $e^{\varepsilon t}$  only. The last one determines the equality of expression under integration when  $t \rightarrow -\infty$ , that means the regularization of the integral. The index  $\varepsilon$  is omitted for short notation.

The chemical potential  $\mu(x)$  depends on the temperature and pressure  $\mu(T(x), P(x))$ . At constant pressure  $\nabla \mu(x) = \frac{\partial \mu}{\partial T} \nabla T(x)$ , and consequently

$$\rho = Q^{-1} \exp \left\{ - \int \beta(x) (H(x) - \mu(x)n(x)) dx - \int_{-\infty}^0 \int e^{\varepsilon t} \mathbf{j}_Q(x; t) \cdot \frac{1}{T^2(x)} \nabla T(x) dt dx \right\}, \quad (18)$$

where

$$\mathbf{j}_Q(x; t) = \mathbf{j}_H(x; t) - \mathbf{j}(x; t) \left( \mu - T \frac{\partial \mu}{\partial T} \right) \quad (19)$$

is the operator of heat flux.

The stationary density of heat flux in the linear approximation with respect to the temperature gradient is

$$\bar{\mathbf{j}}(x) = - \int L_{Q,Q}(x, x') \frac{1}{T^2(x')} \nabla T(x') dx', \quad (20)$$

where

$$L_{Q,Q}(x, x') = \frac{1}{3\beta} \int_0^\beta \int_{-\infty}^0 e^{\varepsilon t} \left\langle \mathbf{j}_Q(x) \left( \mathbf{j}_Q(x'; t + i\hbar\tau) - \langle \mathbf{j}_Q(x') \rangle_q \right) \right\rangle_q d\tau dt \quad (21)$$

is the Green-Cubo formula for the kernel of heat conductivity transport where parameter  $\varepsilon \rightarrow +0$  after the thermodynamical limit. It  $\hbar \rightarrow 0$  (21) transforms into time averaged correlation function of space and time

$$L_{Q,Q}(x, x') = \frac{1}{3} \int_{-\infty}^0 e^{\varepsilon t} \left\langle j_Q(x) \left( j_Q(x'; t) - \langle j_Q(x') \rangle_q \right) \right\rangle_q dt. \quad (22)$$

The heat flux (20) is related to the local entropy production

$$\sigma(x) = \int \left( \nabla \frac{1}{T(x)} \cdot \nabla \frac{1}{T(x')} \right) L_{Q,Q'}(x, x') \cong \lambda(x) (\nabla T(x))^2, \quad (23)$$

where

$$\lambda(x) = \frac{1}{T^2(x)} \int L_{Q,Q'}(x, x') dx' \quad (24)$$

is the heat conductivity coefficient.

Thus if to choose retarding local integrals of motion (12), the local entropy production can't be negative, as it is well-known from the nonequilibrium thermodynamics [12].

The method considered above for construction of the nonequilibrium statistical operator can be easily generalized for non-stationary case. The simplest way is to formulate the selection of needed retarding solution immediately by inserting into the Liouville equation the term depending on the infinitesimal parameter  $\varepsilon \rightarrow +0$ , which breaks the symmetry of equation with respect to the reversal of time. It may be done in two versions.

The first version of the nonequilibrium statistical operator method proceeds from the Liouville equation for logarithm of the statistical operator:

$$\frac{\partial}{\partial t} \ln \varrho(t) + \frac{1}{i\hbar} [\ln \varrho(t), H] = 0, \quad (25)$$

where in the right-hand part the term  $-\varepsilon (\ln \varrho(t) - \ln \varrho_q(t))$  is introduced that breaks the equation's symmetry with respect to the reversal of time. This term is equal zero at  $\varrho(t) = \varrho_q(t)$ ,  $\varepsilon \rightarrow +0$  after the thermodynamical transition when to estimate averages

$$\frac{\partial}{\partial t} \ln \varrho(t) + \frac{1}{i\hbar} [\ln \varrho(t), H] = -\varepsilon (\ln \varrho(t) - \ln \varrho_q(t)). \quad (26)$$

This version of the nonequilibrium statistical operator method is similar to the method of McLennan [14] who considered the contribution into equation (25) of entropy flux, which flows outside from the system on influence of interaction with the environment, these interaction forces being non-potential ones. Perhaps the nonequilibrium statistical operator method is simpler as it doesn't need an obvious introduction of the thermostat replacing it by a boundary condition.

Let's write down the equation (26) in the form

$$\frac{d}{dt} (e^{\varepsilon t} \ln \varrho_\varepsilon(t, t)) = \varepsilon e^{\varepsilon t} \ln \varrho_q(t, t), \quad (27)$$



where

$$\ln \varrho_\varepsilon(t, t) = e^{-\frac{Ht}{i\hbar}} \ln \varrho_\varepsilon(t, 0) e^{\frac{HT}{i\hbar}}, \quad \ln \varrho_q(t, t) = e^{-\frac{Ht}{i\hbar}} \ln \varrho_q(t, 0) e^{\frac{HT}{i\hbar}}.$$

Integrating this equation in the limit  $-\infty$  and  $t$  and supposing that  $\lim_{t \rightarrow -\infty} \ln \varrho_q(t, t) = 0$  we obtain

$$\begin{aligned} \ln \varrho_\varepsilon(t, t) &= \varepsilon \int_{-\infty}^t e^{\varepsilon(t_1-t)} \ln \varrho_q(t_1, t_1) dt_1 \\ &= \varepsilon \int_{-\infty}^0 e^{\varepsilon t_1} \ln \varrho_q(t + t_1, t + t_1) dt_1. \end{aligned} \quad (28)$$

Hence the unknown nonequilibrium statistical operator has the form

$$\begin{aligned} \varrho_\varepsilon(t) = \varrho_\varepsilon(t, 0) &= \exp \left\{ \overset{\sim\sim\sim\sim\sim H}{\ln \varrho_q(t, 0)} \right\} \\ &= \exp \left\{ \varepsilon \int_{-\infty}^0 e^{\varepsilon t_1} \ln \varrho_q(t + t_1, t_1) dt_1 \right\}, \end{aligned} \quad (29)$$

where the wave line from above with the index  $H$  means the operation of extraction of the operator invariant part  $\ln \varrho_q(t, 0)$  relatively to the evolution with Hamiltonian  $H$  moreover  $\varepsilon \rightarrow +0$  after performance of the thermodynamical limit at calculation of operator averages.

We have  $\overset{\sim\sim\sim H}{f(H)} = f(H)$  evidently for any function of the Hamiltonian  $H$ . The Gibbs distribution for the real gas of interacting particles is an invariant part of the Gibbs distribution for the perfect gas that was shown by Prigogine [15] for a quantum case. He named the result, to a certain extent conventionally, the  $H$ -theorem.

The state of the system with given mean values  $\langle P_m(x) \rangle^t$  corresponds to the quasi-equilibrium statistical operator

$$\varrho_q(t) = \exp \left\{ \Phi(t) - \sum_m \int \mathcal{F}_m(x; t) P_m(x) dx \right\}, \quad (30)$$

where

$$\Phi(t) = \ln \text{Sp} \exp \left\{ - \sum_m \int \mathcal{F}_m(x; t) P_m(x) dx \right\}, \quad (31)$$

is the Massier-Plank function,  $\mathcal{F}(x; t)$  are parameters, conjugated with mean values

$$\langle P_m(x) \rangle^t = \text{Sp} (\varrho(t) P_m(x)) \quad (32)$$

and defined from the self-consistency conditions

$$\langle P_m(x) \rangle_q = \langle P_m(x) \rangle^t = \text{Sp} (\varrho(t) P_m(x)), \quad (33)$$

where  $\varrho(t)$  satisfies the Liouville equation.

Quasi-equilibrium statistical operator (30) under additional conditions (33) leads to thermodynamical equalities for  $\Phi(t)$  and entropy

$$S(t) = -\langle \ln \varrho_q \rangle_q = -\langle \ln \varrho_q \rangle^t. \quad (34)$$

Really, we have

$$\begin{aligned} \frac{\delta \Phi(t)}{\delta \mathcal{F}_m(x; t)} &= -\langle P_m(x) \rangle_q = -\langle P_m(x) \rangle^t, \\ \frac{\delta S(t)}{\delta \langle P_m(x) \rangle^t} &= \mathcal{F}_m(x; t), \end{aligned} \quad (35)$$

i.e. the parameters  $\mathcal{F}_m(x, t)$  and  $\langle P_m(x) \rangle^t$  are conjugated.

For a multicomponent gas or liquid

$$P_0(x) = H(x), \quad P_1(x) = \mathbf{P}(x), \quad P_{i+1}(x) = n_i(x), \quad (i \geq 1), \quad (36)$$

where  $H(x)$ ,  $\mathbf{P}(x)$ ,  $n_i(x)$  are the densities of energy, momentum and particle number of components respectively. They are related to the conjugated parameters

$$\begin{aligned} \mathcal{F}_0(x; t) &= \beta(x; t), \\ \mathcal{F}_1(x; t) &= -\beta(x; t)\mathbf{v}(x; t), \\ \mathcal{F}_{i+1}(x; t) &= \beta(x; t)\left(\mu_i(x; t) - \frac{m_i}{2}v^2(x; t)\right), \end{aligned} \quad (37)$$

where  $m_i$  is mass of particles,  $\mathbf{v}(x, t)$  is mass velocity.

This choice of parameters corresponds to the consideration in the coordinate system at which the physically small liquid element moves with mass velocity  $\mathbf{v}(x; t)$

$$\varrho_q(t) = \exp \left\{ -\Phi(t) - \sum_i \int \beta(x; t) \left( H'(x) - \mu_i(x; t)n_i(x) \right) dx \right\}, \quad (38)$$

where

$$H'(x) = H(x) - \mathbf{v}(x; t) \cdot \mathbf{P}(x) + \frac{1}{2}\varrho(x)v^2(x; t) \quad (39)$$

is energy density in moving coordinate system,

$$\varrho(x) = \sum_i m_i n_i(x) \quad (40)$$

is mass density, that is independent of system's motion.

From the extreme condition of Massier-Plank function with respect to variation of mass velocity

$$\frac{\delta \Phi(x)}{\delta \mathbf{v}(x; t)} = 0, \quad (41)$$

we obtain, that

$$\mathbf{v}(x; t) = \frac{\langle \mathbf{P}(x) \rangle^t}{\langle \varrho(x) \rangle^t}, \quad (42)$$

i.e.  $\mathbf{v}(x, t)$  is really the mass velocity.

Quasi-equilibrium distribution (30) corresponds to the operator

$$S(t) = -\ln \varrho_q(t) = \Phi(t) + \sum_m \int \mathcal{F}_m(x; t) P_m(x) dx, \quad (43)$$

the mean value of which is equal to entropy

$$\langle S(t) \rangle^t = \Phi(t) + \sum_m \int \mathcal{F}_m(x; t) \langle P_m(x) \rangle^t dx \quad (44)$$

and this is why it may be called an entropy operator.

Let us represent the nonequilibrium statistical operator (29) in the form

$$\varrho_\varepsilon(t) = \exp \left\{ \overset{\sim}{\sim}{\sim}{\sim} H \right\} = \exp \left\{ -S(t, 0) + \int_{-\infty}^0 e^{\varepsilon t'} \dot{S}(t + t', t') dt' \right\}, \quad (45)$$

where  $S(t) = S(t, 0)$  and the operator

$$\begin{aligned} \dot{S}(t) &= \dot{S}(t, 0) = \frac{\partial}{\partial t} S(t, 0) + \frac{1}{i\hbar} [S(t, 0), H] = \\ &= \sum_m \int \left( \dot{\mathcal{F}}_m(x; t) (P_m(x) - \langle P_m(x) \rangle^t) + \mathcal{F}_m(x; t) \dot{P}_m(x) \right) dx \end{aligned} \quad (46)$$

may be called an operator of the entropy production. Really

$$\langle \dot{S}(t) \rangle^t = \sum_m \int \mathcal{F}_m(x; t) \langle \dot{P}_m(x) \rangle^t dx. \quad (47)$$

It is convenient to write down the operator (46) in the form

$$\begin{aligned} \dot{S}(t) &= \sum_m \int \left( \dot{\mathcal{F}}_m(x; t) (P_m(x) - \langle P_m(x) \rangle_q) + \right. \\ &\quad \left. + \mathcal{F}_m(x; t) (\dot{P}_m(x) - \langle \dot{P}_m(x) \rangle_q) \right) dx, \end{aligned} \quad (48)$$

where we used the identity

$$\langle [\ln \varrho_q, H] \rangle_q = 0. \quad (49)$$

Values  $P_m(x)$  are equal to densities of motion integrals, hence

$$\dot{P}_m(x) = -\nabla \cdot j_m(x), \quad (50)$$

where  $j_m(x)$  are the vector or tensor operators of flux densities. Taking into account (50) the formula (48) takes form

$$\begin{aligned} \dot{S}(t) = \sum_m \int \left( \mathcal{F}_m(x; t) \left( P_m(x) - \langle P_m(x) \rangle_q \right) - \nabla \cdot \left( \mathcal{F}_m(x; t) \left( j_m(x) - \langle j_m(x) \rangle_q \right) \right) + \left( j_m(x) - \langle j_m(x) \rangle_q \right) : \nabla \mathcal{F}_m(x; t) \right) dx \end{aligned} \quad (51)$$

and the total tensor convolution or scalar vector product.

Mean value of the operator of total entropy production is equal

$$\begin{aligned} \langle \dot{S}(t) \rangle^t = - \sum_m \int \nabla \left( \mathcal{F}_m(x; t) \left( \langle j_m(x) \rangle^t - \langle j_m(x) \rangle_q \right) \right) dx + \\ + \sum_m \int \left( \langle j_m(x) \rangle^t - \langle j_m(x) \rangle_q \right) : \nabla \mathcal{F}_m(x; t) dx \end{aligned} \quad (52)$$

Here the first sum corresponds to the entropy flux through the surface of the system, and the second one corresponds to the entropy production in the system. If one remains the surface integral then  $\langle \dot{S}(t) \rangle^t = 0$ .

The second version of the nonequilibrium statistical operator method proceeds from a usual Liouville equation for the statistical operator

$$\frac{\partial}{\partial t} \varrho(t) + \frac{1}{i\hbar} [\varrho(t), H] = 0, \quad (53)$$

where in the right-hand part the term  $-\varepsilon(\varrho(t) - \varrho_g(t))$  is introduced that breaks the equation's symmetry with respect to the reversal of time. This term goes to zero at  $\varepsilon \rightarrow +0$  after the thermodynamical limit transition

$$\frac{\partial}{\partial t} \varrho_\varepsilon(t) + \frac{1}{i\hbar} [\varrho_\varepsilon(t), H] = -\varepsilon(\varrho(t) - \varrho_g(t)). \quad (54)$$

Keeping the normalization of  $\varrho_\varepsilon(t)$  follows immediately from (54), since  $\text{Sp} \varrho_\varepsilon(t) = \text{Sp} \varrho_g(t) = 1$ . Besides that

$$\frac{d}{dt} \langle P_m(x) \rangle^t = \left\langle \frac{d}{dt} P_m(x) \right\rangle^t = -\nabla \cdot \langle j_m(x) \rangle^t, \quad (55)$$

because of self-consistency conditions (33) which are accepted also for the second version of the nonequilibrium statistical operator

$$\langle P_m(x) \rangle^t = \langle P_m(x) \rangle_q \quad (56)$$

and the right-hand part of the equation (54) doesn't contribute into the equation (55) at any  $\varepsilon$ . This property is not possessed by other operators of densities of physical values  $A(x)$  for which only

$$\lim_{\varepsilon \rightarrow +0} \langle A(x) \rangle^t = \langle A(x) \rangle_q. \quad (57)$$

Let us obtain now an obvious solution to the equation (54). For that we write down it in the form

$$\frac{d}{dt} \left( e^{\varepsilon t} \varrho_{\varepsilon}(t, t) \right) = \varepsilon e^{\varepsilon t} \varrho_q(t, t), \quad (58)$$

where

$$\varrho_{\varepsilon}(t, t) = e^{-\frac{Ht}{\hbar}} \varrho_{\varepsilon}(t, 0) e^{\frac{Ht}{\hbar}}, \quad \varrho_q(t, t) = e^{-\frac{Ht}{\hbar}} \varrho_q(t, 0) e^{\frac{Ht}{\hbar}}.$$

Integrating the equation (58) between the limit  $-\infty$  and  $t$  and supposing that  $\lim_{t \rightarrow -\infty} e^{\varepsilon t} \varrho_q(t, t) = 0$  we obtain

$$\varrho_{\varepsilon}(t, t) = \varepsilon \int_{-\infty}^t e^{\varepsilon(t_1-t)} \varrho_q(t_1, t_1) dt_1 = \varepsilon \int_{-\infty}^0 e^{\varepsilon t_1} \varrho_q(t + t_1, t_1 + t, t_1) dt_1, \quad (59)$$

hence the nonequilibrium statistical operator in this version of the method has the form

$$\varrho_{\varepsilon}(t) = \varrho_{\varepsilon}(t, 0) = \overset{\sim\sim\sim\sim H}{\varrho_q}(t, 0) = \varepsilon \int_{-\infty}^0 e^{\varepsilon t_1} \varrho_q(t + t_1, t_1) dt_1. \quad (60)$$

Explanations of a sense of extraction of the quasi-equilibrium part  $\sim\sim\sim\sim$  were presented above after the formula (29). In the formula (60)  $\varepsilon \rightarrow +0$  after the thermodynamical limit transition, and dependence of parameters of a quasiequilibrium distribution on time is defined, like in the previous case, from the self-consistency conditions (56).

It is conveniently to write down the nonequilibrium statistical operator (60) with the help of integration by parts in the form

$$\varrho_{\varepsilon}(t) = \varrho_q(t) + \int_{-\infty}^0 dt' e^{\varepsilon t'} \int_0^1 d\tau e^{-\tau S(t+t', t')} \dot{S}(t+t', t') e^{(\tau-1)S(t+t', t)}, \quad (61)$$

where  $S(t, 0)$  is the operator of entropy production (51).

This version of the nonequilibrium statistical operator method was proposed in papers by D. N. Zubarev and V. P. Kalashnikov [16] and by D. N. Zubarev [17]. It is convenient for exact formulation of nonequilibrium transport equations by the operation of projection of operators separating their slow and rapid part and for the elimination of derivatives by time of parameters  $\mathcal{F}_m(x; t)$  to be considered below. The operation of projection was used to construct equations of the generalized hydrodynamics in papers by S.V.Tishchenko [18] and by M.V.Sergeyev [19].

The proof of equivalence of two methods of the nonequilibrium statistical operator was given by S.V.Tishchenko [20] and by M.I.Auslender [21]. The theory of nonequilibrium processes using the projection method was developed in papers by Zwanzig [22], Robertson [23] and Mori [24].

The Liouville equation (53) one can write in the form

$$\frac{\partial}{\partial t} \varrho(t) + iL\varrho(t) = 0, \quad (62)$$

where  $L$  is a linear self-conjugated Liouville operator, defined by

$$iL\rho = \frac{1}{i\hbar}[\rho, H] \quad (63)$$

(Poisson's quantum bracket).

In case of the classic mechanics Poisson's quantum bracket should be replaced by Poisson's classic one

$$iL\rho = \{\rho, H\} = \sum_k \left( \frac{\partial \rho}{\partial q_k} \frac{\partial H}{\partial P_k} - \frac{\partial \rho}{\partial P_k} \frac{\partial H}{\partial q_k} \right). \quad (64)$$

Convenience of representation of the Liouville equation in the form (62) is connected with its analog with Shrödinger equation.

Equation (54) in the Liouville representation takes form

$$\frac{\partial}{\partial t} \rho_\varepsilon(t) + iL\rho_\varepsilon(t) = -\varepsilon(\rho_\varepsilon(t) - \rho_q(t)), \quad (65)$$

one can rewrite in the form

$$\frac{d}{dt} \left( e^{\varepsilon t} e^{iLt} \rho_\varepsilon(t, 0) \right) = \varepsilon e^{\varepsilon t} e^{iLt} \rho_q(t, 0). \quad (66)$$

Integrating the equation in limits  $-\infty$  and  $t$  and supposing that  $\lim_{t \rightarrow -\infty} e^{\varepsilon t} e^{iLt} \rho_g(t, 0) = 0$  we obtain

$$\begin{aligned} \rho_\varepsilon(t, 0) &= \varepsilon \int_{-\infty}^0 e^{\varepsilon t_1} e^{iLt_1} \rho_q(t + t_1, 0) dt_1 = \\ &= \rho_q(t) - \int_{-\infty}^0 e^{\varepsilon t_1} e^{iLt_1} \left( \frac{\partial}{\partial t_1} \rho_q(t + t_1, 0) + iL\rho_q(t + t_1, 0) \right) dt_1. \end{aligned} \quad (67)$$

Evidently, this expression differs from (61) in only notation form.

Elimination of time derivatives of  $\rho_g(t)$  with the help of projection operators.

To obtain a full equation system for  $\langle P_m(x) \rangle^t$  at any deviation from an equilibrium it is convenient to use the projection operator method.

Let us pass to Fourier-transforms of operators

$$P_{mk} = \int e^{-i(kx)} P_m(x) dx, \quad (68)$$

where  $P_{mk} = P_{m, -k}^+$  since  $P_m(x)$  are real. Let  $n$  means a set of indices  $(m, k)$ ,  $P_k \equiv P_{mk}$ . We accept for future that index  $n$  incorporates index  $m$  determining the type of integral and wave number  $k$ .

Let us write down the Liouville equation with infinitesimal source in the form

$$\left( \frac{\partial}{\partial t} + iL + \varepsilon \right) = - \left( \frac{\partial}{\partial t} + iL \right) \rho_q(t), \quad (69)$$

where  $\Delta\rho(t) = \rho(t) - \rho_g(t)$  is a deviation of the nonequilibrium distribution from the quasiequilibrium one.

Operator  $\rho_q(t)$  depends on time via mean values of  $\langle P_n \rangle^t$  only or via conjugated operators  $\mathcal{F}_n(t)$ . Hence

$$\begin{aligned} \frac{\partial}{\partial t} \rho_q(t) &= \sum_n \frac{\delta \rho_q(t)}{\delta \langle P_n \rangle^t} \frac{\partial}{\partial t} \langle P_n \rangle^t = \\ &= - \sum_n \frac{\delta \rho_q(t)}{\delta \langle P_n \rangle^t} \left\{ \text{Sp} \left( P_n iL \Delta\rho(t) \right) + \text{Sp} \left( P_n iL \rho_q(t) \right) \right\}. \end{aligned} \quad (70)$$

Kawasaki and Gunton proposed to use for elimination of time derivatives by time the projection operator

$$\mathcal{P}_q(t)A = \rho_q(t) \text{Sp}A + \sum_n \frac{\delta \rho_q(t)}{\delta \langle P_n \rangle^t} \left( \text{Sp}(AP_n) - \text{Sp}A \langle P_n \rangle^t \right), \quad (71)$$

where  $A$  is derivative operator with finite trace or zero one. If  $\text{Sp}A = 0$  the Kawasaki-Gunton projection operator coincides with the Robertson one [23]

$$\mathcal{P}_R(t)A = \sum_n \text{Sp}(AP_n) \frac{\delta \rho_q(t)}{\delta \langle P_n \rangle^t}. \quad (72)$$

The Kawasaki-Gunton projection operator is convenient because it, on contrary to the Robertson operator, transfers a nonequilibrium distribution  $\rho(t)$  to a quasiequilibrium one

$$\mathcal{P}_q(t)\rho(t) = \rho_q(t) \quad (73)$$

and has the next properties

$$\mathcal{P}_q(t)\mathcal{P}_q(t') = \mathcal{P}_q(t), \quad \mathcal{P}_q(t) \frac{\partial \rho(t)}{\partial t} = \frac{\partial}{\partial t} \rho_q(t). \quad (74)$$

By (71) we transform the Liouville equation (69) to the form

$$\left( \frac{\partial}{\partial t} + (1 - \mathcal{P}_q(t))iL + \varepsilon \right) \Delta\rho(t) = -(1 - \mathcal{P}_q(t))iL\rho_q(t). \quad (75)$$

Solving this equation with the initial condition  $\lim_{t \rightarrow -\infty} \Delta\rho(t) = 0$  we obtain

$$\Delta\rho(t) = - \int_{-\infty}^t e^{\varepsilon(t'-t)} U(t, t') (1 - \mathcal{P}_q(t')) iL\rho_q(t') dt', \quad (76)$$

where

$$U(t, t') = \exp_+ \left\{ - \int_{t'}^t (1 - \mathcal{P}_q(S)) iL dS \right\}, \quad t \geq t' \quad (77)$$

is ordered exponent.

To obtain equation system for  $\langle P_n \rangle^t$  one has to evaluate the right-hand part of equation

$$\frac{\partial}{\partial t} \langle P_n \rangle^t = S_p \left( \dot{P}_n \varrho(t) \right), \quad (78)$$

where  $\varrho(t)$  is the statistical operator defined by formula (76). Substituting  $\varrho(t)$  from (76) to (78) we find the system of nonlinear transport equations

$$\begin{aligned} \frac{d}{dt} \langle P_n \rangle^t &= \langle \dot{P}_n \rangle_q + \sum_m \int_{-\infty}^0 dt_1 e^{\varepsilon t_1} \mathcal{F}_m(t+t_1) \times \\ &\times \left\langle I_n(t) U(t, t+t_1) \int_0^1 d\tau \varrho_q^\tau(t+t_1) I_m(t+t_1) \varrho_q^{-\tau}(t+t_1) \right\rangle_q, \end{aligned} \quad (79)$$

where  $I_n(t) = (1 - \mathcal{P}(t)) \dot{P}_n$  are flux operators,  $\mathcal{P}(t)$  is Mori projection operator [24], defined by the relation

$$\begin{aligned} \mathcal{P}(t)A &= \langle A \rangle_q - \sum_{m,n} \frac{\delta \mathcal{F}_n(t)}{\delta \langle P_m \rangle^t} (A, P_m) (P_m - \langle P_n \rangle^t) = \\ &= \langle A \rangle_q + \sum_n \frac{\delta \langle A \rangle_q}{\delta \langle P_n \rangle^t} (P_n - \langle P_n \rangle^t) \end{aligned} \quad (80)$$

which projects any operator  $A$  on the space of operators  $P_n$ . Really

$$\mathcal{P}(t) = \langle \mathcal{P} \rangle^t + \sum_m \frac{\delta \langle P_m \rangle^t}{\delta \langle P_n \rangle^t} (P_n - \langle P_n \rangle^t) = P_n. \quad (81)$$

Noticing that

$$\frac{\delta \mathcal{F}_n(t)}{\delta \langle P_n \rangle^t} = \frac{\delta^2 \langle S(t) \rangle}{\delta \langle P_n \rangle^t \delta \langle P_m \rangle^t} = -(P, P)_{mn}^{-1}, \quad (82)$$

where  $(P, P)^{-1}$  is the matrix, inverted to

$$(P_n, P_m) = \frac{\delta^2 \Phi(t)}{\delta \mathcal{F}_n(t) \delta \mathcal{F}_m(t)} = \int_0^1 d\tau \left( P_n, \varrho_q^\tau(t) P_m \varrho_q^{-\tau}(t) \right), \quad (83)$$

we can write down Mori projection operator in the form

$$\mathcal{P}(t)A = \langle A \rangle_q + \sum_{m,n} (A, P_m) (P, P)_{mn}^{-1} (P_n - \langle P_n \rangle^t). \quad (84)$$

Mori considered a particular case of action of projection operator (81) on hydrodynamical variables - Fourier-transforms of operators of densities for energy, number of particles and momentum  $H_k, n_k, P_k$  for statistical equilibrium system, when in (81) all the averages with quasiequilibrium distribution can be replaced by those with the Gibbs equilibrium distribution which



are equal zero at  $k = 0$ . In this case the action of  $\mathcal{P}$  on a Fourier-transform yields

$$\mathcal{P}A_k = \sum_{mn} (A_k, P_{-k,m})(P_k, P_{-k})_{mn}^{-1} P_{k,m}, \quad (85)$$

evidently, that  $\mathcal{P}P_{k,n} = P_{k,n}$

Equations (79) are exact. They contain the evolution in time not with the evolution operator like in (77) but "reduced evolution" like in (77) and that is why they are very complicated.

Transport equation in linear case are a particular result of the formula (79).

There are a lot of papers at present in which the nonequilibrium statistical operator method is applied to the theory of transport phenomena. (see references in [2], [13]). For example this method was applied to transport phenomena in dielectric crystals [26]. It was also used for liquid crystals [27] to obtain equations of relativistic hydrodynamics and Green-Cubo formulae for corresponding kinetic coefficients [28] as well as for hydrodynamics of non-classic liquids [29] and for hydrodynamics of superfluid  $^4\text{He}_2$  [30]. Besides that the method was applied to various generalizations of Boltzmann kinetic equation [31-34], to the theory of "hot" electrons in semiconductors, when electron distribution function differs essentially from the Maxwellian one [35], to numerical problems of nuclear and electron magnetic resonance [36-38] and to the statistical theory of nucleation, i.e. to the kinetics of new phase center formation [39]. Recently the nonequilibrium statistical operator method was also applied to the theory of nonlinear hydrodynamical fluctuations, when it is naturally to use the Fokker-Plank equation in terms of functional derivatives. The similar approach was applied also in the theory of hydrodynamical turbulence.

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## НЕРІВНОВАЖНИЙ СТАТИСТИЧНИЙ ОПЕРАТОР ЯК УЗАГАЛЬНЕННЯ РОЗПОДІЛУ ГІБСА НА НЕРІВНОВАЖНИЙ ВИПАДОК

Д.М. Зубарев

Описується історія методу нерівноважного статистичного оператора