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Form of Nonequilibrium Statistical Operator, Thermodynamic Flows and Entropy Production

V. V. Ryazanov[1*]

[1]*Institute for Nuclear Research Nat. Acad. of Sci. of Ukraine, 47, Nauky Prosp., Kiev 03068, Ukraine.*

Author's contribution

The only author performed the whole research work. Author VVR wrote the first draft of the paper. Author VVR read and approved the final manuscript"

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ABSTRACT

Nonequilibrium statistical operator (NSO) in the form suggested by Zubarev is represented as an averaging operation of the quasiequilibrium statistical operator over the distribution of the lifetime of the system. The form of the density function of the system lifetime affects all its non-equilibrium characteristics. In general, we consider the situation when the distribution density of the system lifetime depends on the current time moment. In the expressions for the fluxes and entropy production additional terms appear in comparison to the expressions derived from Zubarev's NSO. These additional terms can be obtained by applying the principle of maximum entropy.

Keywords: Nonequilibrium statistical operator; distribution of the lifetime; fluxes and entropy production.

1. INTRODUCTION

One of the most fruitful and successful ways of development of the description of the non-equilibrium phenomena is given by a method of the non-equilibrium statistical operator (*NSO*) [1,2,3]. In work [4] new interpretation of the NSO method is given, where the operation of taking the invariant part [1,2,3] or the use of the auxiliary «weight function» (in terminology of [5,6]) in *NSO* are treated as the averaging of the quasi-equilibrium statistical

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Corresponding author: Email: vryazan@yandex.ru;

operator on the distribution of the past lifetime of a system. This approach is consistent with the approach by Zubarev [2] with *NSO* yielded by averaging over the initial time.

This interpretation of *NSO* gives it physical sense of the account of causality and allocation of a real final time interval in which a given physical system is placed. New interpretation leads to various directions of development of *NSO* method which is compared, for example, with Prigogine's [7] approach, introduction of the operator of internal time, irreversibility at microscopically level.

In [5] a source is introduced in the Liouville equation which gives the modified Liouville operator coinciding with the form of the Liouville equation suggested by Prigogine [7] (the Boltzmann-Prigogine symmetry), when the irreversibility is entered in the theory at the microscopic level. We note that the form of *NSO* by Zubarev in the interpretation of [4] corresponds to the main idea of [7] in which one sets to the distribution function ρ_q which evolves according to the classical mechanics laws, the coarse distribution function $\rho = \Lambda\rho_q$ (Λ is operator) whose evolution is described probabilistically since one perform an averaging with the probability density $p_q(u)$, Λ acts as an integral operator.

In Kirkwood's works [8] it was remarked, that the system state in some time moment depends on all previous evolution of the non-equilibrium processes developing in the system. In [5, 6] it is noted, that different «weight functions» can be chosen. Any consistent form of the lifetime distribution density would give a source term in the general form in dynamic Liouville equation which thus acquires the form considered by Boltzmann and Prigogine [5,6,7], and contains dissipative items.

In Zubarev's works [1-3] the linear form of a source corresponding to the limiting exponential distribution for lifetime is introduced. Other choices of the lifetime distribution density would give more exact analogues of the "collision integrals». The approach in facts introduces an explicit account for the time symmetry violation (introducing the finite lifetime, that is the beginning and the end of a system life cycle) is introduced.

In [9-10] it is shown, in what consequences for non-equilibrium properties of system results change of lifetime distribution of system for systems with final lifetime. In [9-11] various dependence of the probability density of time past life $p_q(u)$ from the age of the system are considered, $u=t-t_0$, t is current time, t_0 is the moment of the birth of the system. In [11] the dependence of $p_q(u,t)$ on the current time moment is considered. In [11] this dependence is chosen in the piecewise continuous form, where the form of the function $p_q(u)$ is different for two time intervals. The general case can be considered choosing the continuous function $p_q(u,t)$ with an additional argument t. This choice is considered in the present paper generally and for specific forms of the function $p_q(u,t)$. We show how the choice of this function affects the physical characteristics of the system, namely, flows and entropy production.

2. NEW INTERPRETATION OF NSO

In [3] the nonequilibrium distribution (or NSO) is written in the form

$$\rho(t)=\frac{1}{t-t_0}\int_{t_0}^{t} exp\,\{-i(t-t')L\}\rho_{rel}(t')dt' \qquad (1)$$

where L is Liouville operator; $iL=-\{H,\rho\}=\Sigma_k\{(\partial H/\partial p_k)(\partial\rho/\partial q_k)-(\partial H/\partial q_k)(\partial\rho/\partial p_k)\}$; H is Hamilton function, z or p_k and q_k are momentum and coordinates of particles; $\{\ldots\}$ is Poisson bracket. The relevant distribution has a form

$$\rho_{rel}(t)=exp\{-\Phi(t)-\sum_{j=1}^{n} F_j(t)P_j(t)\}; \qquad (2)$$

$$\Phi(t)=ln\int dz exp\{-\sum_{j=1}^{n} F_j(t)P_j(z)\}. \qquad (3)$$

The Lagrange multipliers $F_j(t)$ are determined from the self-consistency conditions

$$<P_n>^t=<P_n>^t_{rel}=Sp(\rho_{rel}(t)P_n);$$

$P_m(t)$ are some observable macroscopic quantities, dynamical variables [1-3]. For example, they may be the energy, the number of particles, the momentum, or some other variables.

In [1-3] in taking the limit transition for t-t_0, the Abel's theorem is used and the *NSO* is rewritten as

$$ln\rho(z;t)=lim_{\varepsilon\to 0}\varepsilon\int_{-\infty}^{0} dt' exp\{\varepsilon t'\} ln\rho_{rel}(z;t+t',-t')dt', \qquad (4)$$

where $\rho_{rel}(t_1,t_2)=e^{-iLt_2}\rho_{rel}(t_1,0)$. The Liouville equation has a source term

$$\partial\rho/\partial t+iL\rho(t)=-\varepsilon(\rho(t)-\rho_q(t,0)), \qquad (5)$$

which tends to zero ($\varepsilon\to 0$) after the thermodynamic limiting transition. Equation (5) thus possesses the Boltzmann-Bogoliubov-Prigogine symmetry. For (1)

$$\partial\rho/\partial t+iL\rho(t)= (\rho(t)-\rho_q(t,0))/(t-t_0).$$

The statistical distribution before taking limit is

$$ln\rho(z;t)=\varepsilon\int_{-\infty}^{t} dt' exp\{-\varepsilon(t-t')\} ln\rho_{rel}(z;t',t-t')dt'. \qquad (6)$$

In [4, 9-11] the distributions (4), (6) are rewritten as

$$ln\rho(t)=\int_0^{\infty} p_q(y) ln\rho_{rel}(t-y,-y)dy=ln\rho_{rel}(t,0) - \int_0^{\infty} (\int p_q(y)dy)(dln\rho_{rel}(t-y,-y)/dy)dy\ , \qquad (7)$$

where probability distribution density function $p_q(y)$ is interpreted as the lifetime distribution $y=t-t_0$ of the system. We obtain the distribution of (1) from the expression (7) when using a uniform distribution of the form

$$p_q(y)=\begin{cases}\frac{1}{b-a}, & x\in[a,b]\\ 0, & x\notin[a,b]\end{cases},\quad b=t,\quad a=t_0 \tag{8}$$

without using the Abel's theorem. If

$$p_q(y)=\varepsilon\exp\{-\varepsilon y\};\ \varepsilon=1/T=<\Gamma>^{-1}, \tag{9}$$

the expression (7) reduces to the form of the *NSO* [1, 2].

Thus the operations of taking the invariant part [1], averaging over initial conditions [2], temporal coarse-graining [8], choose of the direction of time [5,21] are replaced by averaging on the lifetime distribution. The logarithm of *NSO* (1) is equal to the average from the logarithm of the relevant distribution (2) over the system lifetime distribution. As in [22] we make some estimations about the values P_j. The problem of estimation corresponds to assuming some information about values P_j. Lets assume, that this information consists in assumptions about the finiteness of the system lifetime and about exponential distribution $p_q(y)=\varepsilon\exp\{-\varepsilon y\}$. We shall note that for the logarithm of the nonequilibrium distribution $\ln\rho(t)$, given by equality (7), the equation (5) is valid (after replacement $\partial/\partial t$ by $-\partial/\partial y$ and partial integration of the rhs of (5) it is equal to $d\ln\rho(t)/dt$). The initial conditions $\rho(t_0)=\rho_q(t_0,0)$ [2] are satisfied, if in (7) we assume that $\ln\rho(t_0-y,-y)=0$ at $y>0$, as at the moment of time, smaller than t_0, the system does not exist.

Besides the Zubarev's form of *NSO* [1-3], *NSO* in the Green-Mori form [23] is known, where one assumes the auxiliary weight function [5] to be equal to $W(t,t`)=1-(t-t`)/t$; $w(t,t`)=dW(t,t`)/dt`=1/t$; $\tau=t-t_0$. After averaging one sets $\tau\to\infty$. This situation at $p_q(u)=w(t,t`)$ coincides with the uniform lifetime distribution (8). In [1] this form of *NSO* is compared to the Zubarev's form.

It is possible to specify many specific expressions for the lifetime distribution of system, each of which can possess its advantages. Each of these expressions corresponds to the specific form of the source term in the Liouville equation for the nonequilibrium statistical operator. Generally for $p_q(y)$ this source term has the form

$$J=p_q(0)\ln\rho_q(t,0)+\int_0^\infty(\partial p_q(y)/\partial y)(\ln\rho_q(t-y,-y))dy. \tag{10}$$

If the function $p_q(y,t)$ depends on t as well, the form of the source changes. The Liouville equation holds, for example, under the conditions indicated in [1-3], when $t-t_0\to\infty, \varepsilon\to 0$. Another possibility to arrive at the Liouville equation with zero source is to find a function $p_q(y,t)$ that satisfies the condition $J=0$. To do this it is necessary to solve the integral equation. Setting the form of the function $p_q(u)$ reflects not only the internal properties of the system, but also the impact of the environment on the open system, its characteristics of the interaction with the environment. In [2] a physical interpretation of the function $p_q(u)$ in the form of the exponential distribution is given as a free evolution of an isolated system governed by the Liouville operator. In addition, the system undergoes random transitions whereas the corresponding representing point in the phase space switches from one phase trajectory to another with exponential probability under the influence of a "thermostat", the random time intervals between consecutive switches growing infinitely. This occurs if the parameter of the exponential distribution tends to infinity after taking thermodynamic limit. But real physical systems are finite-sized. The exponential distribution is suitable for the description of completely random systems. The impact of the environment on a system can have more organized character, for example, for a system in the stationary nonequilibrium state with input and output fluxes; so different can be the interaction between the system and environment, therefore various forms of the function $p_q(u)$ different from the exponential form can be set.

One could name many examples of explicitly defining the function $p_q(u)$. Every definition implies some specific form of the source term *J* in the Liouville equation, some specific form of the modified Liouville operator and *NSO*. Thus the family of *NSO* is defined. If the distribution $p_q(u)$ contains *n* parameters, it is possible to write *n* equations for their expression through the parameters of the system. From the other side, they are expressed through the moments of the lifetime. There is the problem of the best choice of the function $p_q(u)$ and *NSO*. In [24] to determine the type of function $p_q(u)$ the principle of maximum entropy for the evolution equations with the source is used.

One can make various assumptions about the form of the function $p_q(u)$, yielding different expressions for the source in the Liouville equation and the behaviour of the nonequilibrium system. The main difference of this paper from [4, 9-11] and expressions (1), (7) is that the function $p_q(u)$ is replaced by the function $p_q(u, t)$, as $p^t_q(y)$ in [19].

3. ADDITIONAL TERMS IN THE EXPRESSIONS FOR THE FLUXES AND ENTROPY PRODUCTION

If instead of the function class $p_q(u)$ the dependence on $p_q(u, t)$ is considered, this results in the change of the Liouville equation for *NSO* $\rho(t)$. In [2-3] expression for $\Delta\rho(t)=\rho(t)-\rho_q(t)$ is obtained in the form

$$\Delta\rho(t)=-\int_{-\infty}^{t} e^{-\varepsilon(t-t')}U_q(t,t')Q_q(t')iL\rho_q(t')dt' , \tag{11}$$

where $U_q(t,t')=\exp\{-\int_{t'}^{t} Q_q(\tau)d\tau\}$, $Q_q=1-\mathrm{P}_q$ is the operator, additional to the Kawasaki-Gunton projection operator. The action of the latter on the quantum or classical variable *A* is defined by

$$\mathrm{P}_q(t)A=\rho_q(t)TrA+\sum_n\{Tr(AP_n)-(TrA)\langle P_n\rangle^t\}\frac{\delta\rho_q(t)}{\delta\langle P_n\rangle^t},$$

$Tr(...)$ is the operation of taking the trace [3]. The operation $Tr(...)$ can be interpreted as the integration over the phase space of *N* particles with subsequent summation over all *N* [3]. For the case of dependence $p_q(u,t)$ instead of (11) we obtain

$$\Delta\rho(t)=\int_{-\infty}^{t}e^{p_q(0,t)(t'-t)}U_q(t,t')\{-Q_q(t')iL\rho_q(t')+\int_0^{\infty}\Delta p_q(u,t')\rho_q(t'-u,-u)du\}dt', \quad (12)$$

where $\Delta p_q(u,t)=p_q(0,t)p_q(u,t)+\frac{\partial p_q(u,t)}{\partial u}+\frac{\partial p_q(u,t)}{\partial t}$. In comparison with [4, 9-11] an additional term $\frac{\partial p_q(u,t)}{\partial t}$ appears.

We obtain an expression for the fluxes

$$\frac{\partial\langle P_m\rangle^t}{\partial t}=\langle\dot{P}_m\rangle^t{}_q+\sum_n\int_{-\infty}^{t}e^{-p_q(0,t')(t-t')}[\mathrm{M}_{mn}(t,t')F_n(t')+\Delta_m(t,t')]dt', \quad (13)$$

where the first term in square brackets is obtained in [2,3]

$$\mathrm{M}_{mn}(t,t')=\int_0^1 dxTr\{I_m(t)U_q(t,t')\rho_q{}^x(t')I_n(t')\rho_q{}^{1-x}(t')\}, \quad (14)$$

$I_n(t)=Q(t)\dot{P}_n+(1-P(t))\dot{P}_n$ are dynamic variables of flows, *P(t)* is Mori projection operator acting on the classical and quantum dynamical variables on the rule $P(t)A=\langle A\rangle^t{}_q+\sum_n\frac{\delta\langle A\rangle^t{}_q}{\delta\langle P_n\rangle^t}(P_n-\langle P_n\rangle^t)$, and the second term presents a correction to the expression obtained in [2, 3]. The appearance of such an additive caused a general form of the density function of the lifetime distribution. In this case,

$$\Delta_m(t,t')=\int_0^{\infty}[p_q(0,t')p_q(u,t')+\frac{\partial p_q(u,t')}{\partial u}+\frac{\partial p_q(u,t')}{\partial t'}]Tr\{I_m(t)U_q(t,t')\rho_q(t'-u,-u)\}du. \quad (15)$$

For $p_q(u)$ in exponential form (10) $\Delta p_q=0$ and, therefore, the addition of (15) is zero. The expression for the entropy production with an additional term in comparison with the expressions derived in [2,3] is:

$$\frac{dS(t)}{dt}=\sum_{m,n}\int_{-\infty}^{t}e^{-p_q(0,t')(t-t')}F_m(t)[\mathrm{M}_{mn}(t,t')F_n(t')+\Delta_m(t,t')]dt'. \quad (16)$$

4. ESTIMATES OF THE ADDITIONAL TERMS

To estimate the magnitude of the addition terms in terms of flows and entropy production we use the explicit expression for the function $p_q(u,t)$ obtained in [24] with the maximum entropy method. Under certain approximations the expression for the distribution of the lifetime obtained in [24] can be written as

$$p_q(u,t) = \frac{p_q(0)e^{-c_i u/F_i}}{1 + \frac{p_q(0)}{F_i} e^{-c_i u/F_i} (R(t) - R(t_0))}, \tag{17}$$

$$R(t) = \sum_j \sum_m F_m(t_0) F_j(t_0) \sum_k \frac{< P_k P_j P_m > - < P_j P_m >< P_k >}{< P_i P_k > - < P_i >< P_k >} + F_i \ln Z(t_0) -$$

$$- \sum_m \sum_j F_j(t_0) \frac{< P_j P_m > - < P_j >< P_m >}{< P_i P_m > - < P_i >< P_m >}, \tag{18}$$

where we use the Zubarev-Peletminsky rule [1,5,25,26]

$$\vec{w}\vec{\nabla}P_i = \sum_{j=1}^{M} C_{ij} P_j, \quad (i = 1,..., M), \quad \frac{d\vec{z}}{dt} = \vec{w}(\vec{z}), \tag{19}$$

where C_{ij} are *c*-numbers. When considering the local density of the dynamical variables, P_i values may depend on the spatial variables. Then the quantities C_{ij} may also depend on the spatial variables or may be differential operators;

$$C_i = \sum_j C_{ji} F_j(t_{\mathbf{0}}). \tag{20}$$

From the normalization condition we find

$$p_q(0) = F_i(1 - e^{-rC_i/F_i^2})/r; \quad r = R(t_0) - R(t).$$

For the distribution of (17) the expression $\Delta p_q(u,t)$, appearing in (15), is

$$\Delta p_q(u,t) = p_q(u,t)[p_q(0,t) - \frac{C_i}{F_i} - \frac{p_q(u,t)}{F_i}(\frac{C_i r}{F_i} - \frac{\partial r}{\partial t})].$$

The value $(\frac{C_i}{F_i})^{-1}$ is close to the average lifetime $\langle t-t_0\rangle$, and following approximate equality can be written: $\frac{C_i r}{F_i}-\frac{\partial r}{\partial t}\approx\frac{r}{\langle t-t_0\rangle}-\frac{\partial r}{\partial t}$. If the value r quickly changes with time this expression can take large values.

In the linear approximation in *r*

$$p_q(0,t_0)=p_q(0)=a=C_i/F_i;\quad p_q(u,t)=ae^{-au}(1+\frac{ar}{F_i}e^{-au});\quad \Delta p_q=\frac{a^2e^{-2ua}}{F_i}(-ar+\frac{\partial r}{\partial t})$$

5. CONCLUSION

Our main result is that, for a specific example of defining a function $p_q(u,t)$ shows the effect of this function on the physical characteristics of the system: flows and entropy production.

In [16-19] the lifetimes of a system are considered as functionals of a random process, that is the random moment for a stochastic process that characterizes the system, to achieve a certain threshold, such as zero level. This definition is used in the present work. In [11, 27-28] the lifetime is included within the range of common physical quantities acting as controls (in terms of information theory) for the quasi-equilibrium statistical operator, and providing additional information about the system. The distribution containing lifetime as thermodynamic parameter considered in [11, 28] can be related to the interpretation of NSO from [4] and in the present paper as an average over the distribution of the lifetime of the system.

Let's notice, that in a case when the value $d\ln\rho_q(t-y,-y)/dy$ (the operator of entropy production σ [1]) in the second item of the right part (9) does not depend on y and is taken out from the integration on y, this second term becomes $\sigma<\Gamma>$, and the expression (9) does not depend on form of the function $p_q(y)$. It is the case, for example, of $\rho_q(t)\sim exp\{-\sigma t\}$, $\sigma=const$. In [29] such distribution is obtained from the principle of maximum entropy with inclusion of the average values of fluxes as constrains.

The form of the density distribution of the lifetime is essential for the kind of expressions for nonequilibrium system behaviour. A more detailed description $p_q(u)$ compared with the limiting exponential (10) allows to describe the real stages of the evolution (and systems with small lifetimes). Each of the distributions for the lifetime has a certain physical meaning. In the queuing theory different service policies correspond to different expressions for the density distribution of a lifetime. In the stochastic theory of storage specifying these expressions corresponds to setting different models of the output and input into the system.

It is shown that the account for dependence of this function on the current point in time leads to additional terms in the expressions for the average flows, of entropy production and other characteristics of a nonequilibrium system.

If the type of source in the Liouville equation for a non-equilibrium statistical operator is chosen in the form suggested by Zubarev [2] it is possible to compare it with the linear relaxation source in the Boltzmann equation; more complicated types of sources from other distributions for lifetime of the system, can be compared to more realistic types of collision integrals. Different forms appear to be representation of the openness of the system, its interaction with surroundings, finiteness of its lifetime, and coarsening procedures for physically infinitely small volumes.

In [30] it was noted that the role of the form of the source term in the Liouville equation in *NSO* method has never been investigated. In [19] it is stated that the exponential distribution is the only one which possesses the Markovian property of the absence of the after action, that is whatever is the actual age of a system, the remaining time does not depend on the past and has the same distribution as the lifetime itself.

The physical sense of averaging over the introduced lifetime distribution of quasi-equilibrium system consists in the obvious account of breaking the time symmetry and information loss related to it, which is manifested in the average of entropy production $<\Delta S\ (t)>$ not equal to zero, obviously reflecting fluctuation-dissipative processes as irreversible phenomena in non-equilibrium systems. The correlations obtained in the present paper generalize the results of statistical non-equilibrium thermodynamics [1, 2 ,3] and information statistical thermodynamics [4-5] as instead of weight function in a form $\varepsilon exp\{\varepsilon t'\}$ the probability density of the lifetime distribution for quasi-equilibrium system is introduced which need not be in exponential form (in the latter case it coincides with weight function from [1, 2, 3]). For example, for system with *n* classes of ergodic states the limiting exponential distribution is replaced with the generalized Erlang function. In the study of lifetimes for complex systems it is possible to involve many results of the theory of reliability, the theory of queues, the stochastic theory of storage processes, theory of Markov renewal, the theory of semi-Markov processes etc.

As it is specified in [31], the existence of time scales and information stream from slower to faster degrees of freedom creates irreversibility of the macroscopical description. The information continuously passes from slow to fast degrees of freedom, which leads to irreversibility. The information thus is not lost, and passes into the form inaccessible to retrieval on the Markovian level of the description. For example, for the rarefied gas the information is transferred from one-partial observables to multipartial correlations. In [4] the values $\varepsilon=1\ /\ <\Gamma>$ and $p_q\ (u)\ =\ \varepsilon exp\ \{-\varepsilon u\}$ are expressed through the operator of entropy production and, according to [31], through the information flow from relevant to irrelevant degrees of freedom.

The introduction of the function $p_q(u)$ into *NSO* corresponds to the specification of the description by means of the effective account of interaction with irrelevant degrees of freedom. In the present work it is shown, how it is possible to expand the description of memory effects within the limits of method *NSO*, to a more detailed account of influence on the system evolution of quickly varying variables through the specified and expanded kind of the lifetime distribution function density.

In many physical problems the finiteness of lifetime can be neglected. Then $\varepsilon\sim 1/<\Gamma> \to 0$. For example, for the case of drops evaporation in a liquid it is possible to show [32], that non-equilibrium characteristics depend on $exp\{y^2\}$; $y=\varepsilon/(2\lambda_2)^{1/2}$, λ_2 is the second moment of correlation function of the fluxes averaged over quasi-equilibrium distribution. Estimations show, that even at the minimum values of lifetime of drops (generally of finite size) and the maximum values ε is the value of $y=\varepsilon/(2\lambda_2)^{1/2}\leq 10^{-5}$. Therefore finiteness of values $<\Gamma>$ and ε does not influence the behaviour of system and it is possible to set $\varepsilon=0$. However in some situations it is necessary to consider finiteness of lifetime $<\Gamma>$ and values $\varepsilon>0$. For example, for nanodrops the effect of finiteness of their lifetime should be already taken into account. For the lifetime of neutrons in a nuclear reactor in [4] the equation for $\varepsilon = 1/<\Gamma>$ is obtained which solution leads to the expression for the average lifetime of neutrons which coincides with the so-called period of a reactor. In [33] the account for the finiteness of lifetime of neutrons result to the corrections to the distribution of neutrons energy.

COMPETING INTERESTS

Author has declared that no competing interests exist.

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