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T.S. VLASENKO,[1] V.M. SYSOEV[2]

[1] Institute for Safety Problems of Nuclear Power Plants, Nat. Acad. of Sci. of Ukraine
(12, Lysogirs'ka Str., Kyiv 03028, Ukraine; e-mail: sonychko@bigmir.net)
[2] Taras Shevchenko National University of Kyiv
(4, Academician Glushkov Prosp., Kyiv 03187, Ukraine)

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INFLUENCE OF EXTERNAL FACTORS ON THE STRUCTURE OF A TWO-COMPONENT FLUID IN A STATIONARY STATE

A method has been proposed to calculate the influence of radiation on the structure of a two-component fluid. A corresponding system of integro-differential equations for the pair correlation function, which are similar to the second equations in the BBGKY chain but with a different effective temperature, is obtained. The usual integro-differential equations for the pair correlation functions with effective temperatures that are determined with the use of the nonequilibrium distribution function for particle momenta are shown to be applicable to the calculation of changes in the structural and thermodynamic properties of irradiated fluids.

Keywords: BBGKY chain, integro-differential equations, nonequilibrium steady state, pair correlation function, effective temperature.

1. Introduction

Let us analyze theoretically the influence of external factors on a fluid system in the thermodynamically equilibrium state. Those factors are assumed to be uniformly distributed in the space. For instance, it may be an aqueous solution of the $\mathrm{RaCl_2}$ salt, in which the salt ions are uniformly distributed over the water volume. Simultaneously, radium ions are sources of α-, β-, and γ-radiation. Therefore, the system is undergone a stationary uniform irradiation. Another example of such a model is a chemical nuclear reaction in a homogeneous system with uniformly distributed radicals. In the cases given above, the Maxwellian distribution of particles in the system changes [1], i.e. the system transits into a nonequilibrium state and tends to return to the equilibrium one. Examples illustrating the changes in the Maxwellian distribution function are shown in Figs. 1 and 2.

In the course of fluid irradiation, active particles insert the negative entropy, which is compensated by the entropy produced by the system itself [2–4]. Hence, it is of interest to examine the corresponding changes in the structural properties of the fluid, in particular, the pair distribution functions.

2. Theoretical Model

The Bogolyubov chain of equations for the space-time-dependent distribution functions $F_n(\mathbf{r},\mathbf{p},t)$ looks like [5]

$$\frac{\partial F_n(\mathbf{r}_1^i,\mathbf{r}_2^j,\mathbf{r}_3^k,\mathbf{p}_1^i,\mathbf{p}_2^j,\mathbf{p}_3^k,t)}{\partial t}=$$
$$=[H^{(n)},F_n(\mathbf{r}_1^i,\mathbf{r}_2^j,\mathbf{r}_3^k,\mathbf{p}_1^i,\mathbf{p}_2^j,\mathbf{p}_3^k,t)]+$$
$$+\rho\sum_{i=1}^{n}\int\frac{\partial\Phi(|\mathbf{r}_i-\mathbf{r}_{n+1}|)}{\partial\mathbf{r}_i}\times$$
$$\times\frac{\partial F_{n+1}(\mathbf{r}_1^i,\mathbf{r}_2^j,\mathbf{r}_3^k,\mathbf{p}_1^i,\mathbf{p}_2^j,\mathbf{p}_3^k,t)}{\partial\mathbf{p}_i}d\mathbf{r}_{n+1}d\mathbf{p}_{n+1},\qquad(1)$$

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where $H^{(n)}$ is the Hamilton function for a closed system with n particles; $F_n(\mathbf{r}_1^i,\mathbf{r}_2^j,\mathbf{r}_3^k,\mathbf{p}_1^i,\mathbf{p}_2^j,\mathbf{p}_3^k,t)$ is the n-th nonequilibrium distribution function depending on the spatial coordinates $\mathbf{r}_1^i$, $\mathbf{r}_2^j$, and $\mathbf{r}_3^k$, momenta $\mathbf{p}_1^i$, $\mathbf{p}_2^j$, and $\mathbf{p}_3^k$, and time t; $\Phi(|\mathbf{r}_i-\mathbf{r}_{n+1}|)$ is the interaction potential between structural elements, and $\rho=\frac{N}{V}$ is the fluid density. It is evident that, in the equilibrium case, Eq. (1) has to be reduced to the well-known equation for an equilibrium pair distribution function $F_2(\mathbf{r},\mathbf{p},t)$, with the help of which the thermodynamic properties of the system at equilibrium can be calculated.

In the stationary equilibrium case, the function $F_2(\mathbf{r},\mathbf{p},t)$ can be factorized as a product of the distribution functions over the coordinates, $F_2(\mathbf{r})$, and momenta, $F_2(\mathbf{p})$ [6]. In the equilibrium case, the distribution function over the momenta, $F(p)$, is the Maxwellian one, and the problem becomes trivial. It is of interest to study the behavior of $F_2(\mathbf{r})$ in the case where a modified Maxwellian distribution $F(p)$ describes an open system that is in a stationary nonequilibrium state under external irradiation.

As a model system, let us consider a two-component fluid [7]. This choice can easily be generalized to a multicomponent fluid system, with the one-component system being only a special case [8].

In order to describe the structural characteristics of fluid systems in the general case, it is enough to know the time-independent pair distribution function $F_2(\mathbf{r}_1,\mathbf{r}_2)$, where $\mathbf{r}_1$ and $\mathbf{r}_2$ are the radius vector of the centers of molecules. With the help of this distribution function, it is not only possible to describe the structural properties of fluids (the coordination number; the average distances between atoms, molecules, and ions; the characteristic correlation length), but also their thermal properties. Hence, for a wide class of problems, the chain of equations (1) can be truncated at the second equation. There are a lot of various methods of determination of the pair distribution function: experimental, theoretical, and computer simulation ones [9, 10]. However, the majority of known methods allow this function to be calculated for a system in the equilibrium state and are not suitable for the description of the system in a stationary nonequilibrium state. This work aims at finding $F_2(\mathbf{r}_1,\mathbf{r}_2)$ for a momentum distribution distorted by an external influence on the fluid system.

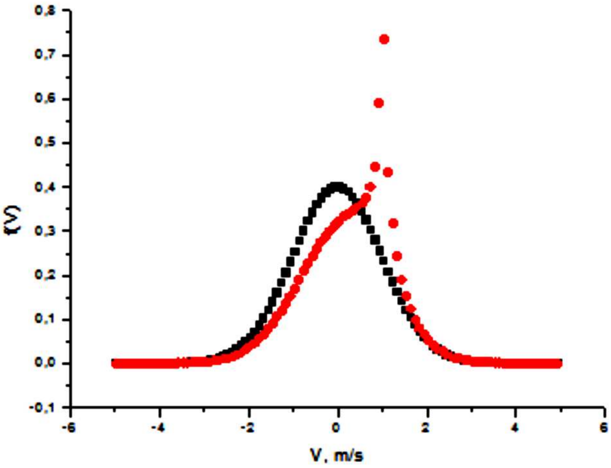

Fig. 1. Variation of the x-component of the particle velocity in a medium under irradiation: the particle velocity before (●) and after (○) the irradiation

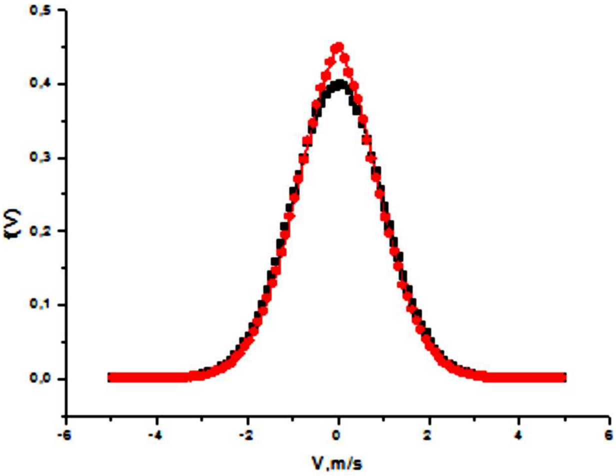

Fig. 2. The same as in Fig. 1, but for the y-component of the particle velocity

The second equation of the Bogolyubov chain (1) for a two-component system in a steady state $\left(\frac{\partial F_n(\mathbf{r}_1^i,\mathbf{r}_2^j,\mathbf{r}_3^k,\mathbf{p}_1^i,\mathbf{p}_2^j,\mathbf{p}_3^k,t)}{\partial t}=0\right)$ looks like

$$0=\left(\frac{\partial\Phi(|\mathbf{r}_1^i-\mathbf{r}_2^j|)}{\partial\mathbf{r}_1^i}\frac{\partial F_2(\mathbf{r}_1^i,\mathbf{r}_2^j,\mathbf{p}_1^i,\mathbf{p}_2^j,t)}{\partial\mathbf{p}_1^i}-\right.$$
$$\left.-\frac{\mathbf{p}_1^i}{m_i}\frac{\partial F_2(\mathbf{r}_1^i,\mathbf{r}_2^j,\mathbf{p}_1^i,\mathbf{p}_2^j,t)}{\partial\mathbf{r}_1^i}\right)+$$
$$+\left(\frac{\partial\Phi(|\mathbf{r}_1^i-\mathbf{r}_2^j|)}{\partial\mathbf{r}_2^j}\frac{\partial F_2(\mathbf{r}_1^i,\mathbf{r}_2^j,\mathbf{p}_1^i,\mathbf{p}_2^j,t)}{\partial\mathbf{p}_2^j}-\right.$$

$$\begin{aligned}
&-\frac{\mathbf{p}_2^j}{m_j}\frac{\partial F_2(\mathbf{r}_1^i,\mathbf{r}_2^j,\mathbf{p}_1^i,\mathbf{p}_2^j,t)}{\partial \mathbf{r}_2^j}\Bigg)+\\
&+\rho_k\sum_{i=1}^{n}\int\limits_{i,j,k=1,2}\frac{\partial\Phi(|\mathbf{r}_1^i-\mathbf{r}_3^k|)}{\partial \mathbf{r}_1^i}\times\\
&\times\frac{\partial F_3(\mathbf{r}_1^i,\mathbf{r}_2^j,\mathbf{r}_3^k,\mathbf{p}_1^i,\mathbf{p}_2^j,\mathbf{p}_3^k,t)}{\partial \mathbf{p}_1^i}d\mathbf{r}_3^k d\mathbf{p}_3^k+\\
&+\rho_k\sum_{i=1}^{n}\int\limits_{i,j,k=1,2}\frac{\partial\Phi(|\mathbf{r}_2^j-\mathbf{r}_3^k|)}{\partial \mathbf{r}_2^j}\times\\
&\times\frac{\partial F_3(\mathbf{r}_1^i,\mathbf{r}_2^j,\mathbf{r}_3^k,\mathbf{p}_1^i,\mathbf{p}_2^j,\mathbf{p}_3^k,t)}{\partial \mathbf{p}_2^j}d\mathbf{r}_3^k d\mathbf{p}_3^k,
\end{aligned}\tag{2}$$

where m_i is the mass of a structural element. Really, in the stationary case, Eq. (2) transforms into a sum of two equations. Provided that the factorization condition $F_2(\mathbf{r},\mathbf{p}) = g_2(\mathbf{r})f_2(\mathbf{p})$ [6] is satisfied, each of them is an equation of the Bogolyubov chain in the case of nonequilibrium state:

$$\begin{aligned}
&\Bigg(\frac{\partial\Phi(|\mathbf{r}_1^i-\mathbf{r}_2^j|)}{\partial \mathbf{r}_1^i}\frac{\partial f_2(\mathbf{p}_1^i,\mathbf{p}_2^j)}{\partial \mathbf{p}_1^i}g_2(\mathbf{r}_1^i,\mathbf{r}_2^j)-\\
&-\frac{\mathbf{p}_1^i}{m_i}\frac{\partial g_2(\mathbf{r}_1^i,\mathbf{r}_2^j)}{\partial \mathbf{r}_1^i}f_2(\mathbf{p}_1^i,\mathbf{p}_2^j)\Bigg)+\\
&+\rho_k\int\limits_{i,j,k=1,2}\frac{\partial\Phi(|\mathbf{r}_1^i-\mathbf{r}_3^k|)}{\partial \mathbf{r}_1^i}\times\\
&\times\frac{\partial f_3(\mathbf{p}_1^i,\mathbf{p}_2^j,\mathbf{p}_3^k)}{\partial \mathbf{p}_1^i}g_3(\mathbf{r}_1^i,\mathbf{r}_2^j,\mathbf{r}_3^k)d\mathbf{r}_3^k d\mathbf{p}_3^k=0,
\end{aligned}\tag{3}$$

$$\begin{aligned}
&\Bigg(\frac{\partial\Phi(|\mathbf{r}_1^i-\mathbf{r}_2^j|)}{\partial \mathbf{r}_2^j}\frac{\partial f_2(\mathbf{p}_1^i,\mathbf{p}_2^j)}{\partial \mathbf{p}_2^j}g_2(\mathbf{r}_1^i,\mathbf{r}_2^j)-\\
&-\frac{\mathbf{p}_2^j}{m_j}\frac{\partial g_2(\mathbf{r}_1^i,\mathbf{r}_2^j)}{\partial \mathbf{r}_2^j}f_2(\mathbf{p}_1^i,\mathbf{p}_2^j)\Bigg)+\\
&+\rho_k\int\limits_{i,j,k=1,2}\frac{\partial\Phi(|\mathbf{r}_2^j-\mathbf{r}_3^k|)}{\partial \mathbf{r}_2^j}\times\\
&\times\frac{\partial f_3(\mathbf{p}_1^i,\mathbf{p}_2^j,\mathbf{p}_3^k)}{\partial \mathbf{p}_2^j}g_3(\mathbf{r}_1^i,\mathbf{r}_2^j,\mathbf{r}_3^k)d\mathbf{r}_3^k d\mathbf{p}_3^k=0,
\end{aligned}\tag{4}$$

where $g_2(\mathbf{r}_1^i)$ and $g_3(\mathbf{r}_1^i)$ are the pair and ternary, respectively, radial distribution functions. Integrating any of those equations, e.g., Eq. (3) over $\mathbf{p}_1^i$ and $\mathbf{p}_2^j$, and making allowance for the relation $\int d\mathbf{p}_3 f_3(\mathbf{p}_1,\mathbf{p}_2,\mathbf{p}_3) = f_2(\mathbf{p}_1,\mathbf{p}_2)$, we obtain a modified BBGKY equation for the case of nonequilibrium stationary state,

$$\begin{aligned}
&-\frac{\partial g_2(\mathbf{r}_1^i,\mathbf{r}_2^j)}{\partial \mathbf{r}_1^i}\int\frac{\mathbf{p}_1^i}{m_i}f_2(\mathbf{p}_1^i,\mathbf{p}_2^j)\partial\mathbf{p}_1^i\partial\mathbf{p}_2^j+\\
&+\frac{\partial\Phi(|\mathbf{r}_1^i-\mathbf{r}_2^j|)}{\partial \mathbf{r}_1^i}g_2(\mathbf{r}_1^i,\mathbf{r}_2^j)\int\frac{\partial f_2(\mathbf{p}_1^i,\mathbf{p}_2^j)}{\partial \mathbf{p}_1^i}\partial\mathbf{p}_1^i\partial\mathbf{p}_2^j+\\
&+\rho_k\int\limits_{i,j,k=1,2}\frac{\partial\Phi(|\mathbf{r}_1^i-\mathbf{r}_3^k|)}{\partial \mathbf{r}_1^i}g_3(\mathbf{r}_1^i,\mathbf{r}_2^j,\mathbf{r}_3^k)d\mathbf{r}_3^k\times\\
&\times\int\frac{\partial f_2(\mathbf{p}_1^i,\mathbf{p}_2^j)}{\partial \mathbf{p}_1^i}\partial\mathbf{p}_1^i\partial\mathbf{p}_2^j=0.
\end{aligned}\tag{5}$$

Dividing this equation by $\int\frac{\partial f_2(\mathbf{p}_1^i,\mathbf{p}_2^j)}{\partial \mathbf{p}_1^i}\partial\mathbf{p}_1^i\partial\mathbf{p}_2^j$, we obtain the following expression

$$\begin{aligned}
&kT_{\rm eff}^{ij}\frac{\partial g_2(\mathbf{r}_1^i,\mathbf{r}_2^j)}{\partial \mathbf{r}_1^i}+\frac{\partial\Phi(|\mathbf{r}_1^i-\mathbf{r}_2^j|)}{\partial \mathbf{r}_1^i}g_2(\mathbf{r}_1^i,\mathbf{r}_2^j)+\\
&+\rho_k\int\limits_{i,j,k=1,2}\frac{\partial\Phi(|\mathbf{r}_1^i-\mathbf{r}_3^k|)}{\partial \mathbf{r}_1^i}g_3(\mathbf{r}_1^i,\mathbf{r}_2^j,\mathbf{r}_3^k)d\mathbf{r}_3^k d\mathbf{p}_3^k,
\end{aligned}\tag{6}$$

where the quantity $kT_{\rm eff}^{ij}$ is defined by the relation

$$\begin{aligned}
&kT_{\rm eff}^{ij}\int\frac{\partial f_2(\mathbf{p}_1^i,\mathbf{p}_2^j)}{\partial \mathbf{p}_1^i}\partial\mathbf{p}_1^i\partial\mathbf{p}_2^j=\\
&=-\int\frac{\mathbf{p}_1^i}{m_i}f_2(\mathbf{p}_1^i,\mathbf{p}_2^j)\partial\mathbf{p}_1^i\partial\mathbf{p}_2^j.
\end{aligned}\tag{7}$$

Relation (7) makes it possible to define the effective temperature of the system in the general case.

It should be noted that, in the case of pair distribution functions, Eq. (7) bring us to an uncertainty of the $\frac{0}{0}$ type. In order to avoid it, let us apply the following procedure. In expression (7), we should calculate the integral over $\mathbf{p}_2^j$ taking advantage of the possibility to factorize the momentum distribution function: $f_2(\mathbf{p}_1^i,\mathbf{p}_2^j) = f_1(\mathbf{p}_1^i)f_1(\mathbf{p}_2^j)$. As a result, we obtain

$$kT_{\rm eff}^{i}\frac{\partial f_1(\mathbf{p}_1^i)}{\partial \mathbf{p}_1^i}=-\frac{\mathbf{p}_1^i}{m_i}f_1(\mathbf{p}_1^i),\tag{8}$$

where $f_1(\mathbf{p}_1^i)$ is the unary distribution function over the momenta. Taking into account that $f_1(\mathbf{p}_1^i)$ is an even function of $\mathbf{p}_1^i$, i.e. $f_1(\mathbf{p}_1) = \psi(p^2)$ and, accordingly, $\frac{\partial f_1(\mathbf{p})}{\partial \mathbf{p}} = 2\mathbf{p}\psi'(p^2)$, after integrating over $\mathbf{p}_1^i$, we obtain the expression

$$kT_{\text{eff}}^i = -\frac{1}{2m}\frac{\int d\mathbf{p}_1^i \psi(\mathbf{p}_1^{i2})}{\int d\mathbf{p}_1^i \psi'(\mathbf{p}_1^{i2})} = -\left(2m\int d\mathbf{p}_1^i \psi'(\mathbf{p}_1^{i2})\right)^{-1}. \tag{9}$$

It is evident that the difference of expression (7) from Eq. (9) consists in the absence of the uncertainty mentioned above. Hence, Eq. (9) makes it possible to calculate the effective temperatures in a fluid system in a stationary nonequilibrium case. One can easily see that if the function $\psi(\mathbf{p}_1^{i2})$ corresponds to the Maxwellian distribution over the momenta, we expectedly obtain $kT_{\text{eff}}^i = kT_{\text{eff}}^j = kT_{\text{eff}}$.

3. Experimental Verification

The main characteristic times used at the description of a fluid structure are the average period of atomic vibrations, τ_0, and the average dwell time of atoms in the equilibrium state, τ ($\tau > \tau_0$). Three types of structures are distinguished on the basis of this hierarchy [11]:

1) an instant structure of the nearest environment of an atom, which is essential for fast processes with characteristic times $t < \tau_0$;

2) the averaged structure of the nearest environment of an atom within the time interval $\tau_0 < t < \tau$; and

3) the averaged structure of the nearest environment of an atom, which is essential at slow and equilibrium processes with characteristic times $t \gg \tau$.

Using the structural classification of fluids, the thermodynamic and physical parameters of the system can be divided into two groups: dependent on the instant or the averaged structure of a fluid. The former includes the electric properties of fluids, whereas the latter their viscosity, thermal conductivity, diffusion, and surface tension.

In this work, the attention is concentrated on an electric property of fluids, namely, on the electric conductivity. Owing to a large difference between the masses of free electrons and atoms, and the resulting difference between their velocities, it is possible to consider the processes supposing the atoms to be at rest. This is the so-called adiabatic approximation. Therefore, the individual motions of separate electrons are governed by the instant structure and *vice versa.*

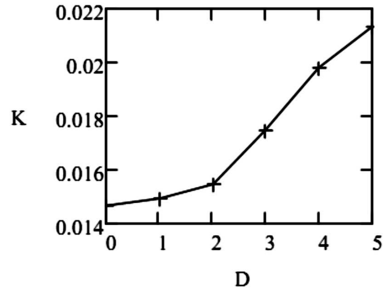

Fig. 3. Dependence of the specific electric conductance in a KCl solution with a concentration of 0.001 M on the γ-radiation exposure dose [12]

Suppose that the stationary nonequilibrium state can be regarded as an infinite sequence of similar instant structures. From Fig. 3, one can see that the specific electric conductance of a KCl solution with a concentration of 0.001 M is equal to $K_0 = 0.015\ \Omega^{-1}\text{m}^{-1}$ before the irradiation, which corresponds to the nonequilibrium system temperature $T_0 = 301$ K, and to $K = 0.0215\ \Omega^{-1}\text{m}^{-1}$ after the irradiation, which corresponds to the temperature $T = 323$ K (see Table 1). Hereafter, the specific electric conductance was evaluated taking its linear concentration dependence into account. Therefore, for the

Table 1. **Temperature dependence of the specific conductance of a KCl solution with a concentration of 0.01 M [13]**

T, °C	K, $\Omega^{-1}\text{m}^{-1}$	T, °C	K, $\Omega^{-1}\text{m}^{-1}$
28	0.1496	42	0.1899
30	0.1552	44	0.1957
32	0.1609	46	0.2015
34	0.1667	48	0.2073
36	0.1725	50	0.2131
38	0.1783	52	0.2189
40	0.1841	54	0.2247

Table 2. **Specific conductance of a KCl solution with a concentration of 1.2 M before and after irradiation [12]**

K, $\Omega^{-1}\text{m}^{-1}$ before irradiation	K, $\Omega^{-1}\text{m}^{-1}$ after irradiation	Specific conductance decrease, %
10.784	10.542	2.24

Table 3. **Temperature dependence of the specific conductance of the KCl solution with a concentration of 1.0 M [13]**

T, °C	K, $\Omega^{-1}\mathrm{m}^{-1}$
12	8.689
13	8.876
14	9.063
15	9.252
16	9.441

diluted KCl solution with a concentration of 0.001 M, the effective temperature equals $kT_{\text{eff}} \approx 1.07\,kT$.

Let us consider a concentrated KCl solution with a concentration of 1.2 M. From Table 2, one can see that its specific conductance is equal to $K_0 = 8.987\ \Omega^{-1}\mathrm{m}^{-1}$ before the irradiation, which corresponds to the temperature $T = 288$ K, and to $K_0 = 8.785\ \Omega^{-1}\mathrm{m}^{-1}$ after the irradiation, which corresponds to the temperature $T = 286.5$ K (Table 3). Hence, for the KCl solution with a concentration of 1.2 M, the effective temperature equals $kT_{\text{eff}} \approx 1.005\,kT$.

From the results obtained, one can see that, in the case of diluted solutions, the effective temperature of the system differs from the measured one more substantially than in the case with the concentrated one. Accordingly, in the framework of the proposed approach, we may say that variations in the instant structure are larger for the system with a lower concentration. Whence a conclusion can be drawn that the distribution of particles over their momenta differs from the Maxwellian one more strongly just for such systems.

This qualitative picture corresponds to physical processes running in the system. For instance, provided identical radiation doses, the numbers of particles with changed velocities at different concentrations will be close to one another. At the same time, the total number of particles taken into consideration in the velocity distribution will be larger. Accordingly, the influence of particles with changed velocities on the general distribution will be lower for systems with higher concentrations.

4. Conclusions

A method has been proposed for the calculation of the pair distribution functions describing an open system in a stationary nonequilibrium state. The developed method showed that the formalism of equilibrium thermodynamics allows the stationary nonequilibrium systems to be described by introducing an effective temperature.

The results obtained testify that the thermodynamic properties of a fluid system under the action of external factors vary as a result of a deviation of the momentum distribution function from the Maxwellian law, which, in turn, gives rise to variations in structural characteristics. As a result, there arise a few effective temperatures in the fluid system, which can be explained by the different actions of the external field on the momentum distribution functions of its subsystems. This situation corresponds to the results of the statistical theory of relaxation processes for systems consisting of weakly interacting subsystems [14].

The analysis of calculation results demonstrates that, in the case of diluted solutions, the effective temperature of the system differs from the measured one more substantially than in the case of concentrated solutions.

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Т.С. Власенко, В.М. Сисоєв

ВПЛИВ ЗОВНІШНІХ ФАКТОРІВ НА СТРУКТУРУ ДВОКОМПОНЕНТНИХ РІДИН В СТАЦІОНАРНОМУ СТАНІ

Резюме

Проведено теоретичне дослідження зовнішніх факторів, вплив яких приводить двокомпонентну рівноважну термодинамічну систему до стаціонарного нерівноважного стану зі скалярними потоками. Отримана система інтегродиференціальних рівнянь для парних кореляційних функцій, що збігаються з другими рівняннями ланцюжка ББГКІ, але з різними ефективними температурами. Таким чином, для розрахунку структурних та термодинамічних властивостей такої системи можна користуватися звичайними інтегродиференціальними рівняннями для парної кореляційної функції з ефективними температурами, що записуються через збурену (нерівноважну) функцію розподілу Максвела за імпульсами.