Relaxation time scales in collective dynamics of liquid alkali metals

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In this paper the investigation of the dynamical processes of liquid alkali metals is executed by analyzing the time scales of relaxation processes in liquids. The obtained theoretical dynamic structure factor \(S(k, \omega)\) for the case of liquid lithium is found to be in excellent agreement with the recently received inelastic x-ray scattering data. The comparison and interrelation with other theories are given here. Finally, an important part of this paper is the confirmation of the scale uniformity of the dynamic processes in liquid alkali metals predicted by some previous molecular dynamic simulation studies. © 2004 American Institute of Physics. [DOI: 10.1063/1.1792155]

I. INTRODUCTION

The dynamic structure factor \(S(k, \omega)\) is an experimentally measured term, containing information about the processes in a liquid with long- and short-time scales. It can be used to judge the microscopic behavior in a system on the basis of its spectra, obtained by means of inelastic neutron scattering (INS) (Refs. 1 and 2) or inelastic x-ray scattering (IXS).\(^3\) As for simple liquids, at present a great amount of experimental data of \(S(k, \omega)\) have been accumulated, in particular, for liquid alkali metals. These data indicate legibly the presence of the collective propagation excitations beyond the hydrodynamic region. The characteristic feature of liquid alkali metals is a triple-peak shape of \(S(k, \omega)\), lasting to \(k \sim 0.8k_m\), where \(k_m\) corresponds to the first maximum of the static structure factor \(S(k)\). Moreover, the frequency of the side peak achieves its maximum at \(k \sim 0.55k_m\). The propagation of these high-frequency waves cannot be obtained within a hydrodynamic treatment, therefore, they are related in some works to the so-called kinetic collective excitations. The impossibility to describe these microscopic phenomena and, therefore, to reproduce qualitatively the experimental \(S(k, \omega)\) by means of ordinary hydrodynamic equations led to the development of other theoretical models and approaches.

One of the simplest and perhaps the earliest modeling approaches is the so-called viscoelastic theory. It allows one to obtain the central quasielastic line as well as two inelastic peaks symmetrically located around \(\omega=0\) for mesoscopic space-frequency region. However, as shown in Refs. 4 and 5, this model cannot be used for the exact reproduction of the experimental spectral shapes of \(S(k, \omega)\) (see, for instance, the cases of liquid cesium and lithium in Refs. 4 and 5). Therefore, in Ref. 5 the double-scale model for the viscous relaxation process with fast and slow time scales was tested, and as a result a good agreement with the IXS experimental data for the dynamic structure factor was received. Recently the similar approach was also applied for the description of relaxation processes in H-bonded liquids.\(^6\)\(^7\) The existence of two time scales in this model reflects the presence of physically different decay mechanisms. A faster process is hypothetically associated with interactions between an atom and the “cage” of its nearest neighbors, and a slower one is identified with the well-known structural \((\alpha-\) ) process. However, relaxations of both processes are approximated by exponential dependencies. In recent works the viscoelastic model has been improved by means of the Markovian closure on the next relaxation level of Zwanzig-Mori hierarchy\(^8\), it is equivalent to the exponential relaxation on this level. It is worth mentioning two other methods, one of which is related to the extension of the usual hydrodynamic analytical expressions by modification of hydrodynamic modes to \(k\) dependence (see, for instance, Ref. 9). This method assumes the existence of nonhydrodynamical additional modes. The second approach is related to the so called concept of generalized collective modes, which was proposed for the investigation of the time correlation functions (TCF’s) beyond the hydrodynamic region.\(^10\) The key idea of this method consists in the correct choice of the basic set of dynamical variables.

All these methods are more or less successfully used for the description of collective dynamics in liquids. They have common property. Namely, they are actually constructed on heuristic assumption about the presence of exponential decay (or combination of exponential decay contributions) in some relaxation processes. Nevertheless, the transition and imposition of different relaxation modes in disorder systems can occur even in case of a concrete relaxation process, which complicates the selection of the analytical time dependence for the corresponding TCF. This fact is proved by the successful application of different mode-coupling theories. On the other hand, this difficulty can be resolved by means of analysis and comparison of the resulting time scales of relaxation processes. Therefore, in the present work we suggest the approach, which allows us to avoid the immediate approximation of relaxation processes by analytical functions. It is based on the development of Bogoliubov’s ideas about the hierarchy of relaxation times in liquids,\(^11\) adapted to the formalism of time correlation functions.

One of the open problems in studying of liquid state (in
particular, of the microdynamics of simple liquids) is to describe and understand on a general ground the common features of different relaxation processes. It is well known that the dispersion of the side (high frequency) peak of dynamic structure factor is the same for all alkali metals. Moreover, it is also valid in case of more complex systems, for example, for liquid alloys. Then the following questions arise: Is the origin of relaxation processes the same for liquid systems with similar features? Can the unified description be applied to these systems? As for the group of melting alkalis, it has been indicated in Ref. 13 that both the equilibrium and the time dependent correlations can be cast in a properly scaled form for all the alkali metals. Further, it was justified by ab initio molecular dynamics studied in Ref. 13 too. Experimental confirmation of this result was impossible over a long period particularly because of the difficulties related to the technique of INS due to the deficient precision of the experimental data. Recently, due to progress in IXS technique this issue was considered again. In this work we present investigations related to the determination of corresponding scale transitions for liquid systems.

The organization of the paper is as follows. In the following section, we describe the theoretical formalism, and the comparison with the experimental data and other theories is carried out. The possibility of scale uniformity of dynamical processes in the group of liquid alkali metals is analyzed and discussed in Sec. III. The scale-crossing relations are also presented here. Finally, we come up with some concluding remarks in Sec. IV.

II. THEORETICAL FORMALISM

A. Basic notions

Let us consider the liquid system of \( N \) identical classical particles of the mass \( m \) in the volume \( V \) and take the density fluctuations

\[
W_0(k) = \frac{1}{\sqrt{N}} \sum_{j=1}^{N} e^{ikr_j}
\]

(1)
as an initial dynamical variable. To construct some set of dynamical variables necessary for the description of the evolution of the system we use the technique of projection operators of Zwanzig-Mori. It is a formal version of the Gram-Schmidt orthogonalization process, which allows one to obtain the set of orthogonal variables,

\[
W(k) = \{W_0(k), W_1(k), W_2(k), \ldots, W_j(k), \ldots\}
\]

(2)
They satisfy the condition \( \langle W^*_n W_j \rangle = \delta_{n,j} \langle |W_j|^2 \rangle \) and are connected by the following recurrent relation:

\[
W_{j+1}(k) = \mathcal{L} W_j(k) - \Omega_j^0(k) W_{j-1}(k),
\]

\( j = 0, 1, 2, \ldots, \)

\( W_{-1}(k) = 0. \)

Here the characteristic of the corresponding \( j \)th relaxation process, the so-called frequency parameter \( \Omega_j^0(k) \), appears, and \( \mathcal{L} \) is the Liouville operator,

\[
\mathcal{L} = -i \left[ \sum_{j=1}^{N} \frac{p_j \nabla_j}{m} - \sum_{i>j=1}^{N} \nabla_j u(j,i)(\nabla_j - \nabla_i) \right],
\]

(4)
with the momentum of the \( j \)th particle \( p_j \) and the pair potential \( u(j,i) \). So, if \( W_0(k) \) is the density fluctuation, then \( W_1(k) \) is the longitudinal component of the momentum density and so it goes on.

The TCF’s for the corresponding dynamical variables are given by

\[
M_{jl}(k,t) = \frac{\langle W_j^*(k)e^{i\mathcal{L}t/2}W_l(k) \rangle}{\langle W_j^*(k)W_l(k) \rangle}, \quad j,l = 1, 2, \ldots.
\]

(5)
For convenience normalized time correlation functions are used here. The time evolution operator of Eq. (5) contains the reduced Liouville operator,

\[
\mathcal{L}^{(i)}_{22}(t) = \left( 1 - \sum_{j=1}^{l} \Pi_j \right) \mathcal{L} \left( 1 - \sum_{j=1}^{l} \Pi_j \right)
\]

(6)
defined by the following projection operators:

\[
\Pi_j = \frac{W_j(k)}{\langle |W_j(k)|^2 \rangle}, \quad \Pi_j \Pi_l = \delta_{j,l} \Pi_j.
\]

(7)
From the condition of orthogonalization of the dynamical variables we obtain the initial values for the TCF’s of Eq. (5)

\[
M_{jl}(k,t=0) = \begin{cases} 0 & \text{if } j \neq l, \text{ cross correlations} \\ 1 & \text{if } j = l, \text{ autocorrelations} \end{cases}
\]

(8)
These correlation functions \( M_{jl}(k,t) \) are symmetrical in \( l \) and \( j \), i.e.,

\[
M_{jl}(k,t) = M_{lj}(k,t).
\]

(9)
Autocorrelation functions of Zwanzig-Mori formalism have the following property: every autocorrelation function of the higher order \( M_{j}(k,t) = M_{jl}(k,t) \) is a memory function for the previous one, i.e., \( M_{j-1}(k,t) \) (autocorrelation functions will be marked by one index only in accordance with the used variable), and they are interrelated by integro-differential non-Markovian equations of the form:

\[
\frac{dM_{j-1}(k,t)}{dt} + \Omega_j^0(k) \int_0^t d\tau M_{j}(k,\tau)M_{j-1}(k,\tau - \tau) = 0.
\]

(10)
Differentiating the first equation of the chain (10), i.e., \( j = 1, \) one obtains the generalized Langevin equation:

\[
\frac{d^2M_0(k,t)}{dt^2} + \Omega_1^0(k)M_0(k,t) - \Omega_1^0(k)\Omega_2^0(k) \int_0^\tau d\tau' \int_0^\tau d\tau'' M_2(k,t-\tau)M_1(k,t-\tau')M_0(k,\tau') = 0.
\]

(11)
One the other hand, these functions describe concrete relaxation processes, the physical meaning of which may be established from direct definitions of TCF’s. For instance, \( M_{0}(k,t) \) describes the dynamics of fluctuations of density correlations in the system, \( M_{1}(k,t) \) is the TCF of the fluctuations of the longitudinal component of the momentum density, and \( M_{2}(k,t) \) contains the TCF of fluctuations of energy density. So, these quantities are associated with the...
TCF’s of the well-known hydrodynamic “slow” variables. These TCF’s have characteristic time scales, which can be found from

\[ \tau_f(k) = \text{Re} \int_0^\infty dt M_f(k, t) = \text{Re} \tilde{M}_f(k, s = 0), \]

where \( \tilde{M}_f(k, s) \) is the Laplace transform of the corresponding TCF, i.e., \( \tilde{M}_f(k, s) = \int_0^\infty dt e^{-st} M_f(k, t) \) (Refs. 18–20).

So, the memory function approach with single initial dynamical variable extracts the whole set, which describes the relaxation processes of the corresponding relaxation levels.

In fact, the well-known problem of the choice of a set of variables required for the correct description of the system dynamics here is reduced (i) to the search of the number of variables for \textit{a priori} known succession \( W(k) \), that was excellently shown by the recurrent relation approach in a works of Lee,21,22 and/or (ii) to finding the correct closure of the chain (10).

The ratio between \( \tau_0(k) \), \( \tau_1(k) \), and \( \tau_2(k) \) may be quite arbitrary. In the hydrodynamic region \((k \to 0, \omega \to 0)\) they take large values due to the slow changes of the correspondent variables: densities of mass, momentum, and energy. Further, one can suggest that the relaxation times of the following TCF’s, in comparison with the scales of these three variables, are comparable, i.e., \( \tau_3(k) \approx \tau_4(k) \). We emphasize here that this assumption does not contradict the viscoelastic model, which presupposes that \( \tau_3(k) \approx \tau_4(k) \). Obviously, this key condition of the viscoelastic theory is just a special case in our approach. Simultaneously, our approach does not deny the presence of the long-lasting time tail of \( \tilde{M}_2(k, t) \), which may be adequately taken into account by the mode-coupling theory.23

Then, taking into account Eq. (12) one can find

\[ M_4(k, t) = M_3(k, t) + h(t), \]

where the “tail” function \( h(t) \) appears. From the short-time asymptotic of the time autocorrelation functions and the condition of the long-time attenuation of correlation Eq. (13) yields the following properties of \( h(t) \):

\[ \lim_{t \to 0} h(t) = \lim_{t \to \infty} h(t) = 0, \]

this function must have at least one crossing with the time axis at the intermediate region.24 Equation (13) allows us to obtain the closure of hierarchy of equations of the form (10) at the fourth level \((j = 4)\) and by means of Laplace transformation to find its exact solution for \( \tilde{M}_0(k, i \omega) \), particularly, which is directly related to the experimentally available term and the dynamic structure factor, \( S(k, \omega) \). The expression for the resulting \( S(k, \omega) \) is given in Ref. 25 in terms of the first four frequency parameters \( \Omega_1^2(k), \Omega_2^2(k), \Omega_3^2(k), \) and \( \Omega_4^2(k) \) and the Laplace transform of tail function, i.e., \( \tilde{h}(k, i \omega) \). In some cases, the regime with \( h(t) \to 0 \) may be realized. It can be observed in some parts of time (frequency) scale. In this case we find the following expression for the dynamic structure factor:

\[
S(k, \omega) = \frac{S(k)}{2\pi} \Omega_1^2(k) \Omega_2^2(k) \Omega_3^2(k) \Omega_4^2(k) [4 \Omega_4^2(k) - \omega^2]^{\nu_1_2} \Omega_2^2(k) \Omega_3^2(k) - 2 \Omega_1^2(k) \Omega_4^2(k) \\
- \Omega_1^2(k) \Omega_2^2(k) + 2 \Omega_1^2(k) \Omega_3^2(k) \Omega_4^2(k) - \Omega_1^2(k) \Omega_2^2(k) \Omega_3^2(k) + \Omega_2^2(k) \Omega_3^2(k) + \omega^2[\Omega_2^2(k) - 2 \Omega_1^2(k) \Omega_4^2(k)] \\
+ 2 \Omega_1^2(k) \Omega_3^2(k) - 2 \Omega_2^2(k) \Omega_3^2(k) + \Omega_4^2(k) \Omega_3^2(k) + \omega^2[\Omega_4^2(k) - 2 \Omega_1^2(k) \Omega_3^2(k)] - 1. \quad (15)
\]

This equation is also expressed through the first four frequency parameters, which are directly related to the first five even frequency moments of dynamics structure factor. It is necessary to note that this expression is obtained in the way completely different from the theory of moments.26

**B. Comparison with IXS experiment and relationship with other theoretical approaches**

In Fig. 1 we report the dynamic structure factor \( S(k, \omega) \) of liquid lithium \((T = 475 K)\) for some wave numbers calculated from Eq. (15) (solid line) and obtained from IXS experiment (circles).14 Being used in theoretical computations the static structure factor \( S(k) \) for both cases was taken from Ref. 27. The first frequency parameter was directly defined from its definition \( \Omega_1^2(k) = K_B T k^2 / m S(k) \). The second frequency parameter \( \Omega_2^2(k) \) is related to the fourth frequency moment. We found this parameter from the values of the infinite frequency sound velocity \( c_\infty(k) \) (Refs. 5 and 14) by means of relation \( c_\infty(k) = \sqrt{\Omega_1^2(k) + \Omega_2^2(k)} / k \). The high-order parameters were found by comparison with the experiment. Eventually, we have revealed that all the frequency parameters have the similar dispersion. In particular, they have the first principal maximum at the same wave numbers such as the side peak of \( S(k, \omega) \), i.e., at \( k \sim 0.55k_m \), and any low order parameter is less than the high order one.

We would like to emphasize that the theoretical \( S(k, \omega) \) and, in particular, the position of the side peak, is very sensitive to the magnitude of \( \Omega_2^2(k) \). The magnitudes of \( \Omega_2^2(k) \) and \( \Omega_3^2(k) \) influence the form of \( S(k, \omega) \). However, it is not so important to know these parameters separately as their ratio, i.e., \( \Omega_2^2(k)/\Omega_3^2(k) \).

To compare the theoretical outcome with the experiment we modified it to account for the quantum mechanical detailed balance condition according to

\[
S_q(k, \omega) = \frac{\hbar \omega / K_B T}{1 - e^{-\hbar \omega / k_B T}} S(k, \omega), \quad (16)
\]
\[ \xi = \frac{s^2}{4 \Omega_4^2}, \quad |\xi| \ll 1. \]  

Taking into account the fact that the found values of \( \Omega_4^2(k) \) for liquid sodium and lithium achieve \( 10^{20} \ldots 10^{30} \text{s}^{-2} \) for the low-\( k \) region, we span by introducing parameter \( \xi \) the frequency (time) range \( \omega \leq 10^{15} \text{s}^{-1} (t \leq 10^{-15} \text{s}) \), which is important for us and is available experimentally.

Expanding the radicand in Eq. (18b) as a series in the parameter \( \xi \)
\[ 1 + \xi = 1 + \frac{\xi}{2} \frac{\xi^2}{8} + \cdots, \]
we can rewrite it in the following way:
\[ \widetilde{M}_\lambda(s) = -\frac{s}{2 \Omega_4^2} + \frac{1}{\Omega_4} + \frac{s^2}{8 \Omega_4^2} - \frac{s^4}{32 \Omega_4^4} + \cdots. \]

By restricting the number of terms in the series (20) [and, accordingly, in Eq. (21)] we receive from Eq. (18a) the linear combination of the Lorentz functions
\[ M_2(k,t) = \sum_j A_j(k) e^{-t/\tau_j(k)}. \]

By restricting the first term of the series (20) only we receive the simplest model from the first equality of Eq. (18a) with Eq. (21)
\[ M_2(k,t) = e^{-t/\tau(k)}, \]
which corresponds to the viscoelastic model with the relaxation time \( \tau(k) = \Omega_4(k)/\Omega_3^2(k) \), and from the second equality of Eq. (18a) the double exponential model, i.e., Eq. (23) at \( j = 2 \) with the following time relaxation parameters
\[ \tau_2(k) = \left[ \Omega_4(k) \pm \sqrt{\Omega_4^2(k) - \Omega_3^2(k)} \right]^{-1} \]
and the weight factor
\[ A(k) = \frac{\Omega_4(k) + \Omega_3^2(k) - \Omega_3^2(k)}{2 \sqrt{\Omega_4^2(k) - \Omega_3^2(k)}}. \]

This case may be related to the double-time viscous model.\(^5\text{,14} \) two-time exponential ansatz.\(^18\text{,29} \) In the general form Eq. (23) corresponds to the framework of generalized collective mode approach with the sum of the weighed exponents for the TCF \( M_2(k,t) \), where \( \tau_j^{-1}(k) \) denote eigenvalues of a generalized dynamic matrix with the elements consisting of static correlation functions, and the weight factors \( A_j(k) \) are the amplitudes describing the contribution of the corresponding modes.
So, it is obvious that the theory underlying Eq. (15) prescribes such behavior of the second-order memory function $M_2(k,t)$, which may be represented in the form of Eq. (23) and can be reduced to the above-mentioned models. Eq. (23) is in fact an expansion of $M_2(k,t)$ into decay channels embedded in this function.

### III. SCALE UNIFORMITY OF DYNAMICS PROCESSES IN LIQUID ALKALI METALS

The determination of the scale uniformity of structural and dynamical features for different groups of liquids is very important for the physics of liquid state. On the one hand, it allows one to apply the unified theoretical description to the whole group. On the other hand, it allows one to remove the difficulties related to obtaining the experimental data. The fact is that until recently the microscopic dynamics of liquids could be experimentally probed by INS only. However, there were often different problems related to, first, separation of collective and one-particle contributions, and, second, gross experimental errors (and even with impossibility to obtain data) for different $(k,\omega)$ regions. Recent progress in the technique of IXS has allowed one to clear some of the obstacles.\(^3\) Ten years ago the possibility of the unified description of the structural and dynamical properties of different liquid alkali metals near the melting point was found by the comprehensive molecular dynamics simulation study,\(^3\) where the adopted potential model of Price, Singwi, and Tosi was used, and the scale passage was executed on the basis of the potential parameters. The recent sketchy attempt of testing this outcome experimentally has shown its inconsistency.\(^4\)

In present work we also execute the comparison of the dynamic structure factor spectra of liquid lithium and sodium. As known from the experimental results, the dynamic structure factor $S(k,\omega)$ depends strongly on the temperature $T$ and the wave number $k$. So, one can define the reduced forms of these terms as $T/T_m$ and $k/k_m$, where $T_m$ is the melting temperature and $k_m$ is the main peak position in the static structure factor $S(k)$ for the corresponding system. The scale time interval $t^*$ can be expressed as $t^* = k^{-1}\sqrt{m/K_B T}$. Thus defined time unit $t^*$ is different from the one introduced in Ref. 14, because the present term varies with the change of space and temperature characteristics. However we do not exclude the possibility that this scale unit may be independent of the temperature and the wave number for other systems (for instance, semiconductors, or H-bonded liquids).

In Fig. 2 we report the comparison of $S(k,\omega)$ spectra for liquid lithium and sodium\(^5\)\(^6\)\(^7\)\(^8\)\(^9\)\(^10\)\(^11\)\(^12\)\(^13\)\(^14\) at approximately the same reduced temperatures $T/T_m$ and wave numbers $k/k_m$. Namely, $T/T_m = 1.049$ for liquid lithium and $1.051$ in case of sodium. From this figure one can see that dynamic structure factor practically coincides in the first two higher cases. From the lower plot of Fig. 2 one can see that the position of inelastic and central peaks for both systems is the same. However, though the overall coincidence of spectra is observed at intermediate frequencies only, the peak altitudes are a little different. Such deviation can easily be explained by the fact that the plot for liquid lithium is presented for a higher value of the reduced wave number, 0.75, whereas in case of sodium, $k/k_m = 0.73$. As known, these wave numbers correspond to the so called de Gennes narrowing region characterized by a strong $k$ dependence. In other words, a higher section at $k$ of flat $S(k,\omega)/t^*$ is presented for lithium than for sodium. It is necessary to take into account that the values of the reduced temperatures for both systems are also slightly different.

Notice that the time unit $t^*$ depends on the system features $m$, on the probed spatial region $k$ and the temperature regime $T$ in contrast to scale units $k_m$ and $T_m$, which remain unchanged and the spatial region and the temperature of the system are revised.

At result, the experimental or theoretical $S(k,\omega)$ for any single metal allows one to easily restore this term for the whole group of alkali metals at the same reduced conditions, $k/k_m$ and $T/T_m$. Moreover, the theory developed for the concrete separate alkali metal may be simply extended to the whole group.

As an example, in Fig. 3 we report the dynamic structure factor of liquid potassium $S^K(k,\omega)$ obtained on the basis IXS data for liquid sodium $S^Na(k,\omega)$. The transition $S^Na(k,\omega) \rightarrow S^K(k,\omega)$ has been executed by means of the following scale reductions:

\[
S^K(k,\omega) = S^Na(k,\omega) \frac{k^Na}{k^K} \sqrt{\frac{m^KNa}{m^NaT^K}}.
\]

\[
\omega^K = \omega^Na \frac{k^K}{k^Na} \frac{m^NaT^K}{m^KNaT^Na}.
\]
IV. CONCLUDING REMARKS

The following results are presented in this work.

(i) The theory, developed on the basis of Bogoliubov ideas about the hierarchy of relaxation times, allows one to obtain dynamic structure factor, reproducing adequately experimental IXS spectra for liquid alkali metals (in particular, for liquid lithium and sodium) in the region of low values of wave number.

(ii) The expansion of the second-order memory function into exponential decay channels, used (sometimes intuitively) in others theories, may be easily obtained within the framework of the presented approach. This is the evidence of the multimode character of decay of the observed relaxation process.

(iii) An important result of this work is the confirmation of the proposition about the unitary description of the dynamical features of liquid alkali metals, and finding of corresponding scale transition relations.

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20 Equation (12) defines square under the function $M_j(k,t)$. It takes large values (is turned to infinity) for slow damping (undamping) processes, and it is finite for ordinary ergodic cases. So, it can be chosen as an estimation measure of the relaxation scale. The main advantage of such definition consists in direct establishing relation to the experiment. For example, $\tau_{G}(k)=\tau_{G}(k)/S(k)$. There are other definitions of relaxation time. For instance, P. Egelstaff and W. Gla¨ ser have shown in Ref. 18 that “time of TCF” may be found from $\tau_{G}(k)=[\sum_{j}^{\infty}M_j(k,t)]t_0d[f^{\infty}_{M_j(k,t)}dt]$. Some ideas about relaxation time are also given in Ref. 19.
22 Via recurrent relations approach was shown that finding, for example, of $M_0(k,t)$ can be reduced to the definition of the dimension of a space formed by the set of basic “vectors” $W_0$, $W_1$, $W_2$,…, $W_j$. For infinite spaces at $j=\infty$ given TCF may be also found at the known parameters $\Omega^2$, $\Omega_2^2$,…, or at the known rule of their intercoupling. Taking into account the fact that $\Omega^2(k)$ contains $j$-th particle distribution functions, the quest for their intercoupling may be reduced to the quest for the way to uncouple many-particle distribution functions. However, even on solving this problem the quest for the final result is complicated by multiplex reckonings.
24 It should not be confused with the tail function of MCT, where it defines the long-time behavior of relaxation, whereas the function $\tau_{h}(k,t)$ is the difference of $M_0(k,t)$ and $M_1(k,t)$.