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## Diffusion processes and memory effects

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**Abstract.** We report the results of the numerical estimation of statistical memory effects in diffusion for two various systems: Lennard-Jones fluids and the model of the Brownian particle in a one-dimensional harmonic lattice. We have found the relation between the diffusion coefficient and the non-Markovity parameter, which is linear for the Lennard-Jones systems in liquid state. The relation between the memory measure and the excess entropy is also discussed here.

The famous Einstein relation between the diffusion coefficient and the long time behaviour of the mean-square displacements of a particle as a function of time [1] is a basis for different modern physical approaches (for example, the fractal Einstein relation [2]), although its derivation by Einstein was originated by a *direct* attempt to describe the irregular motion of particles suspended in a liquid, observed in a microscope by the botanist Robert Brown in 1828. At present, transport phenomena attract a lot of attention due to the investigations on space-time nonlocality and memory effects. Disorder can arise in two different ways. Firstly, the investigated matter can have some settled disorder structure with long-range correlations similar to the fractal geometrical structure. Secondly, disordering of the background originates directly through temporal correlations in interactions between random walkers and the background [3]–[6]. The mathematical language of fractional derivatives [7, 8] and steady distributions [9] have allowed us to generalize and expand the physical theory of stochastic transport. Such anomalous dynamics has been investigated theoretically and observed experimentally in amorphous semiconductors [10], turbulent flows [11], surface diffusion [12], low-dimensional lattice gases [13], polymers and plasmas [14], subrecoil laser cooling [15] and has been successfully described by means of continuous time random walk approaches [7], where the stochastic theory yields non-Gaussian probability density functions exhibiting stretched exponential and Lévy power law decays for anomalous transport. Although intensive studies of normal and anomalous diffusion (super- and

sub-) have been carried out, the role of memory effects in diffusion processes (even for the case of the normal diffusion described by the Einstein relation) induced by disorder of the surroundings is not completely clear [3]–[6], [16]. This work is devoted to the study of influence of memory effects on disordering processes in a system.

It is well known that the diffusion coefficient in a three-dimensional isotropic system is defined by the famous Einstein formula [1, 17]:

$$\langle |\Delta \mathbf{r}(t)|^2 \rangle = 6Dt, \quad t \rightarrow \infty. \quad (1)$$

Then, taking into account that

$$\langle |\Delta \mathbf{r}(t)|^2 \rangle = 2 \int_0^\infty (t - t') \langle v_\alpha(0) v_\alpha(t') \rangle dt', \quad (2)$$

and the mean-squared velocity is  $\langle |v_\alpha(0)|^2 \rangle = 3k_B T/m$ , one can obtain from equation (1) for the velocity autocorrelation function  $a(t) = \langle v_\alpha(0) v_\alpha(t) \rangle / \langle |v_\alpha(0)|^2 \rangle$  the Green–Kubo’s relation for the diffusion constant [18, 19]:

$$D = \frac{k_B T}{m} \int_0^\infty a(t) dt, \quad (3)$$

where  $k_B$ ,  $T$  and  $m$  are the Boltzmann constant, the temperature and the mass of the particle, respectively.

On the other hand, the technique of projection operators and recurrent relation approach allow one to obtain the generalized Langevin equation (GLE) from the Heisenberg equation of motion [4, 20, 21]:

$$\frac{d}{dt} v_\alpha(t) = -\omega^{(2)} \int_0^\infty M_1(t - \tau) v_\alpha(\tau) d\tau + \frac{F(t)}{m}, \quad (4)$$

where  $\omega^{(2)} = \langle |\mathcal{L} v_\alpha|^2 \rangle / \langle |v_\alpha|^2 \rangle$  is the second frequency moment of  $a(t)$  with the Liouville operator  $\mathcal{L}$ , and the normalized memory function  $M_1(t)$  is related to the stochastic force  $F(t)$  by means of the fluctuation–dissipation theorem:

$$\langle F(t) F(0) \rangle = m k_B T \omega^{(2)} M_1(t). \quad (5)$$

Multiplying equation (4) by  $v_\alpha(0) / \langle |v_\alpha(0)|^2 \rangle$  and performing an appropriate ensemble average  $\langle \dots \rangle$ , we obtain the GLE of the following form:

$$\frac{da(t)}{dt} = -\omega^{(2)} \int_0^t M_1(\tau) a(t - \tau) d\tau, \quad \langle v_\alpha(0) F(t) \rangle = 0. \quad (6)$$

In case of short-range memory  $M_1(t) = 2\tau_1 \delta(t)$ , equation (6) is reduced to the ordinary Langevin equation [22]:

$$m \frac{d}{dt} v_\alpha(t) + m \omega^{(2)} \tau_1 v_\alpha(t) = F(t) \quad (7)$$

with the ordinary exponential solution for VACF:

$$a(t) = e^{-\omega^{(2)}\tau_1 t}. \quad (8)$$

Further application of the technique of the projection operators leads to the whole chain of interconnected integro-differential non-Markovian equations of the form (6) containing memory functions  $M_n(t)$  and even frequency moments  $\omega^{(2n)}$ ,  $n = 1, 2, 3, \dots$ :

$$\omega^{(2)} = \frac{4\pi n}{3} \int_0^\infty dr g(r) r^2 \left[ \frac{3}{r} \frac{\partial U(r)}{\partial r} + r \frac{\partial}{\partial r} \left( \frac{\partial U(r)}{r \partial r} \right) \right], \quad (9)$$

where  $n$  is the density,  $g(r)$  the radial distribution function and  $U(r)$  the potential of interparticle interaction.

$$\begin{aligned} \omega^{(4)} = & \frac{8\pi n}{3m} \int_0^\infty dr g(r) \left[ 3 \left( \frac{dU(r)}{dr} \right)^2 + \left( r \frac{\partial}{\partial r} \left( \frac{\partial U(r)}{r \partial r} \right) \right)^2 \right. \\ & \left. + \frac{\partial U(r)}{\partial r} \cdot \frac{\partial}{\partial r} \left( \frac{\partial U(r)}{r \partial r} \right) \right] \\ & + \frac{8\pi^2 n^2}{3m} \int_0^\infty \int_0^\infty dr dr_1 r^2 r_1^2 \int_{-1}^1 d\beta g_3(\mathbf{r}, \mathbf{r}_1) \left[ \frac{3}{rr_1} \frac{\partial U(r)}{\partial r} \cdot \frac{\partial U(r_1)}{\partial r_1} \right. \\ & \left. + \frac{r}{r_1} \frac{\partial U(r_1)}{\partial r_1} \cdot \frac{\partial}{\partial r} \left( \frac{\partial U(r)}{r \partial r} \right) + \frac{r_1}{r} \frac{\partial U(r)}{\partial r} \cdot \frac{\partial}{\partial r_1} \left( \frac{\partial U(r_1)}{r_1 \partial r_1} \right) \right. \\ & \left. + rr_1 \frac{\partial}{\partial r_1} \left( \frac{\partial U(r_1)}{r_1 \partial r_1} \right) \cdot \frac{\partial}{\partial r} \left( \frac{\partial U(r)}{r \partial r} \right) \beta^2 \right], \quad (10) \end{aligned}$$

where  $g_3(\mathbf{r}, \mathbf{r}_1)$  is the distribution function of three particles,  $\beta$  the cosine between  $\mathbf{r}$  and  $\mathbf{r}_1$  (see, for example, [23]). The expression for  $\omega^{(6)}$  also contains quadruplet correlation functions [24]:

$$\begin{aligned} \omega^{(6)} = & \frac{4\pi}{m^3} \int d\mathbf{r} g(\mathbf{r}) (U_{xy}(r)U_{xx}(r)U_{yy}(r) + 3k_B T U_{xyz}^2(r)) \\ & + \frac{n^2}{m^3} \iint d\mathbf{r} d\mathbf{r}_1 g_3(\mathbf{r}, \mathbf{r}_1) [3k_B T U_{xxy}(r)U_{xxy}(r_1)] \\ & + U_{xx}(r) [6U_{xy}(r)U_{xy}(r_1) - U_{xy}(r)U_{xy}(\mathbf{r} - \mathbf{r}_1)] \\ & + \frac{n^3}{m^3} \iiint d\mathbf{r} d\mathbf{r}_1 d\mathbf{r}'_2 g_4(\mathbf{r}, \mathbf{r}_1, \mathbf{r}_2) U_{xx}(r)U_{xy}(r_1)U_{xy}(r'_2), \quad (11) \end{aligned}$$

where

$$U_{xyz}(r) = \frac{\partial^3 U(r)}{\partial r_x \partial r_y \partial r_z},$$

$x, y, z$  are the Cartesian components.

The Laplace transformation reduces this chain of non-Markovian equations to the infinite continued fraction of the following form:

$$\begin{aligned}\tilde{a}(s) &= \int_0^\infty dt e^{-st} a(t) = [s + \omega^{(2)} \tilde{M}_1(s)]^{-1} \\ &= 1/\{s + \omega^{(2)}/[s + (\omega^{(4)}/\omega^{(2)} - \omega^{(2)}) \tilde{M}_2(s)]\} \\ &= 1/\{s + \omega^{(2)}/[s + (\omega^{(4)}/\omega^{(2)} - \omega^{(2)})/(s + \dots)]\},\end{aligned}\quad (12)$$

and equation (3) to the form

$$D = \frac{k_B T}{m} \tilde{a}(s=0). \quad (13)$$

So, the calculation of diffusion constant is reduced to a search for the velocity autocorrelation function and truncation of infinite continued fraction (12).

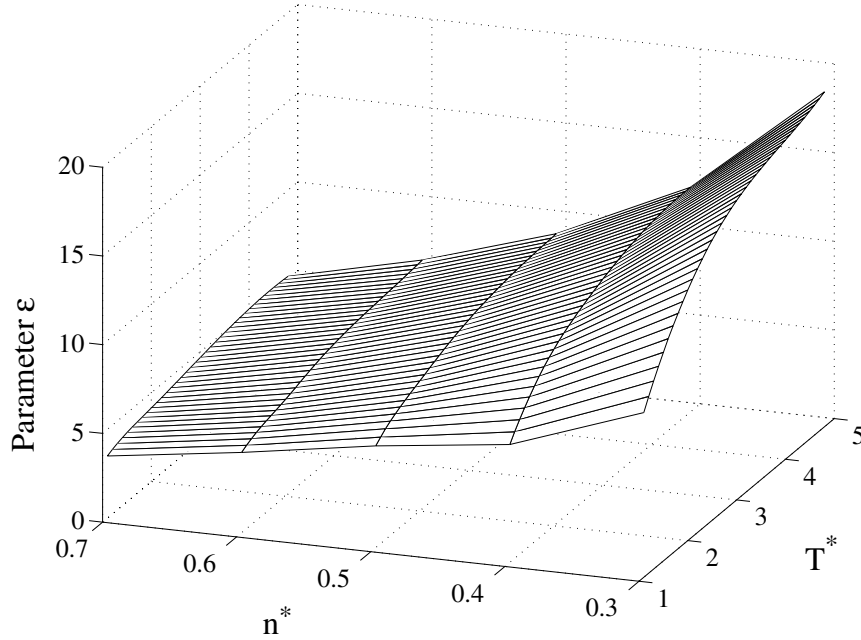
The numerical estimation of the memory effects for diffusion can be executed by a simple comparison of the relaxation scale of VACF,  $\tau_0$ , with the time scale of its memory,  $\tau_1$ , as

$$\varepsilon_0 = \tau_0/\tau_1. \quad (14)$$

The quantity  $\varepsilon_0$  is known as the non-Markovity parameter [25] and may be generalized to  $\varepsilon_n = \tau_n/\tau_{n+1}$ , where  $\tau_n = \int_0^\infty dt e^{-st} M_n(t)$  is the relaxation time of the  $n$ th order memory function  $M_n(t)$ . The expression for  $\varepsilon_0$  may be obtained from equation (14) taking equation (13) and the second equality in equation (12) at  $s = 0$  into account:

$$\varepsilon_0 = \omega^{(2)} \left[ \frac{mD}{k_B T} \right]^2 = \frac{4\pi n}{3} \left[ \frac{mD}{k_B T} \right]^2 \int_0^\infty dr g(r) r^2 \left[ \frac{3}{r} \frac{\partial U(r)}{\partial r} + r \frac{\partial}{\partial r} \left( \frac{\partial U(r)}{r \partial r} \right) \right]. \quad (15)$$

The non-Markovity parameter  $\varepsilon_0$  obtained for a wide range of the temperature  $T^* = k_B T/\epsilon$  and the density  $n^* = n\sigma^3$  of Lennard-Jones fluids is presented in figure 1. Here  $\sigma$  and  $\epsilon$  are parameters of the potential. The values of  $D$  and  $\omega^{(2)}$  were taken from [26, 27] and [28], respectively. As one can see from the figure, the non-Markovity parameter here always satisfies the condition  $\varepsilon_0 > 1$ . This is the evidence of comparatively weak memory effects in diffusion processes, which is usually observed in Markovian and quasi-Markovian processes. In this case, the relaxation time of memory is much smaller than the relaxation scale of the velocity autocorrelation function. The obtained result explains the amazing efficiency of different Markovian approximations in the study of diffusion in Lennard-Jones liquids in the range of low densities and high temperatures [29]. As can be seen from figure 1 the non-Markovity parameter has its minimal values  $\sim 4$  near the triple point with  $n^* = 0.849$  and  $T^* = 0.773$ . This is a quantitative evidence of considerable memory effects in the diffusion processes of Lennard-Jones system in this phase range, characterized by a well-known negative correlation in the velocity autocorrelation function behaviour  $a(t)$ . Furthermore, the parameter  $\varepsilon_0$  smoothly increases with the increase of temperature and decrease of density. In this case Markovization in the movement of particles is observed, which may be related to the rise of disorder in diffusion processes.



**Figure 1.** The density- and the temperature-dependence of the non-Markovity parameter  $\varepsilon$  for VACF of Lennard-Jones systems for the following region of the phase diagram:  $0.3 \leq n^* \leq 0.7$  and  $1 \leq T^* \leq 4.7$ ,  $n^* = n\sigma^3$ ,  $T^* = k_B T/\varepsilon$ , where  $\sigma$  and  $\varepsilon$  are parameters of Lennard-Jones potential.

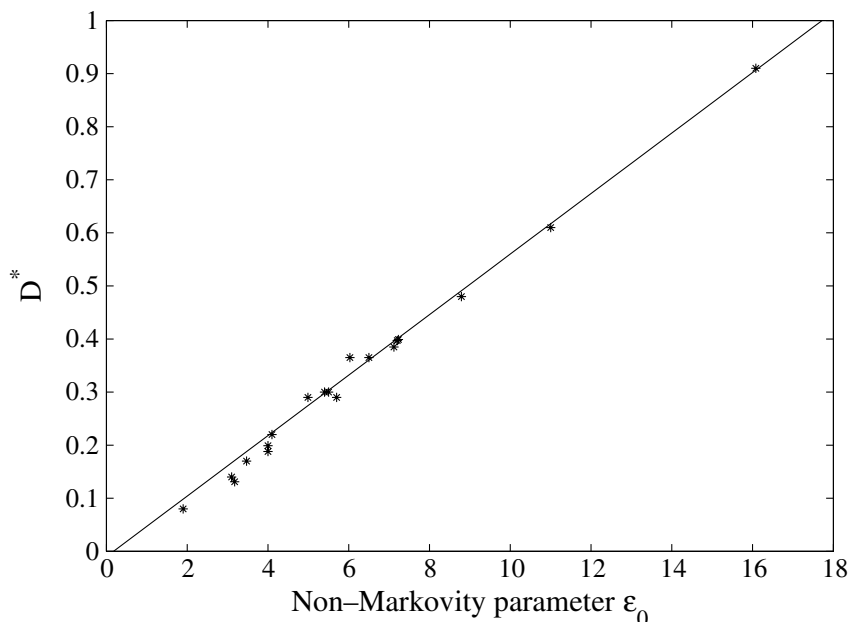
The dependence of the diffusion coefficient on the non-Markovity parameter for the case of Lennard-Jones fluids is presented in figure 2. The calculations are carried out according to equation (15) on the basis of the diffusion results of the molecular dynamics simulations and data of  $\omega^{(2)}$  published in [26]. As can be seen from the figure, the diffusion coefficient in a Lennard-Jones system increases with the increase in the non-Markovity parameter (with the weakening of statistical memory effects). Moreover, the dependence between these quantities is practically linear. The interpolator procedure reveals the following relation:

$$D^* = C\varepsilon_0, \quad C = 0.057. \quad (16)$$

As a result, the more disorder there is in the system, the weaker are the memory effects. From this interpretation, the non-Markovity parameter acquires the quality of a disorder measure comparable with configuration entropy. The direct relation between the non-Markovity parameter and entropy can be received from the scaling relationship between the diffusion coefficient and the excess entropy  $S = (S_{liquid} - S_{id.gas})/k_B$  suggested by Dzугotov [30]. In the original formulation of Dzугotov this relationship has the following form:

$$D^* = \frac{D}{\sigma^2 \Gamma_E} = C_{univ} e^S, \quad (17)$$

where  $\sigma$  is the particle diameter,  $\Gamma_E = 4\sigma^2 g(\sigma)n\sqrt{\pi k_B T/m}$  is the average pair collision frequency of an equivalent hard sphere fluid and the  $C_{univ} = 0.09$  is taken from [31]. Numerous testings of



**Figure 2.** Variation of the diffusion coefficient  $D^* = D(m/\epsilon\sigma^2)^{1/2}$  with the non-Markovity parameter  $\epsilon_0$ . Solid line is the result of linear interpolation.

equation (17) have justified its validity for a model system of hard spheres, for binary mixtures, liquid metals (Ag, Au, Cu, Ni, Pd, Pt) and metallic alloys (Ni<sub>3</sub>Al and AuPt) [30]–[36] in the range of high densities ( $0.68 < n\sigma^3 < 0.936$ ) including the range of supercooled liquids. As for the whole phase range of a liquid state, the coefficient  $D^*$  increases with the rise in  $S$ . However, the exact dependence between these quantities obtained from the initial equations and true of the whole phase range remains a very interesting challenge to pursue [35].

Taking into account equation (15), we obtain from equation (17)

$$\epsilon_0 \sim e^S. \quad (18)$$

Then, for the case of Lennard-Jones liquids we obtain from equations (17) and (16):

$$\epsilon_0 = \frac{C_{univ}}{C} \sigma \Gamma_E \sqrt{\frac{m}{\epsilon}} e^S, \quad (19)$$

where  $C_{univ}/C = 1.57$ . Equations (18) and (19) determine the inter-relationship between the measure of memory effects in the system, the non-Markovity parameter, and the measure of the number of accessible configurations of the system, the excess entropy. However, it is necessary to note that the restriction in equation (17) by the area of high density refers to equation (19) as well. So, if the non-Markovity parameter  $\epsilon_0$  is defined for the velocity autocorrelation function as considered here, it also is the measure of *the system disorder*.

Another part of our study is devoted to the analysis of memory effects and the definition of the non-Markovity parameter for the model of the Brownian motion of Rubin [37], where heavy isotropic impurity ( $M$ ,  $x_0$  and  $v_0$  are the mass, the coordinate and the velocity, respectively) behaves similar to a freely moving Brownian particle with a frictional force proportional to its

velocity. This particle is located in an infinite, one-dimensional harmonic lattice (particles of the lattice have the same mass  $m$ ) with interactions between their nearest neighbours ( $K$  is the spring constant). As a result, the energy of the system is

$$E = \frac{M}{2}v_0^2 + \frac{m}{2}\sum_{j=1}^{N-1}v_j^2 + \frac{K}{2}\sum_{j=0}^{N-1}(x_j - x_{j+1})^2, \quad j = 0, 1, 2, \dots, \quad (20)$$

and the equation of motion of particles is

$$[m + (M - m)\delta_{j,0}]\ddot{x}_j = K(x_{j+1} - 2x_j + x_{j-1}). \quad (21)$$

To carry out the study of the diffusion time scales and to define the diffusion constant of impurity, it is necessary to know the long-time behaviour of the velocity autocorrelation function  $a(t) = \langle v_0(0)v_0(t) \rangle / \langle v_0^2(0) \rangle$ ,  $\langle v_j v_0 \rangle = (k_B T / M)\delta_{j,0}$ ,  $\langle x_j v_0 \rangle = 0$ .

The Laplace transformation  $\tilde{a}(s) = \int_0^\infty dt e^{-st} a(t)$  is defined from equation (21) by its solution with the help of the Laplace transformation and transition to other coordinates (see pp 24–26 in [3]). It has the following form:

$$\tilde{a}(s) = \frac{q}{(q - 1)s + \sqrt{s^2 + 4K/m}}, \quad (22)$$

where  $q$  is the mass ratio:

$$q = \frac{M}{m}. \quad (23)$$

Then, the diffusion constant is

$$\mathcal{D} = \frac{k_B T}{M} \tilde{a}(s = 0) = \frac{k_B T}{2\sqrt{Km}}. \quad (24)$$

The inverse Laplace transformation can be found in two cases: if  $M$  and  $m$  have the same values, i.e.  $q = 1$ ; and if  $q = M/m = 2$ . The solutions for both cases are expressed in terms of the Bessel functions of order 0 and 1, respectively:

$$a(t) = \begin{cases} J_0(2t\sqrt{K/m}), & \text{if } q = 1, \\ \frac{J_1(2t\sqrt{K/m})}{t\sqrt{K/m}}, & \text{if } q = 2. \end{cases} \quad (25)$$

Note that the behaviour of  $a(t)$  at large  $q$  approaches ordinary exponential dependence (except for the small  $t$ ) [37]. It is interesting that the second equation of the system (25) corresponds *exactly* to the approximation  $M_1(t) = a(t)$  for GLE (6).

The short-time asymptotic of  $a(t)$  may be found from the large  $s$  expansion:

$$\tilde{a}(s) = \frac{1}{s} - \frac{2K}{qms^3} + \frac{2K^2(2+q)}{q^2m^2s^5} - \frac{4K^3(q^2+2q+2)}{q^3m^3s^7} + \mathcal{O}(1/s^9). \quad (26)$$



Then one obtains

$$\begin{aligned} a(t) &= 1 - \frac{\omega_a^{(2)}}{2!} t^2 + \frac{\omega_a^{(4)}}{4!} t^4 - \frac{\omega_a^{(6)}}{6!} t^6 + \mathcal{O}(t^8) \\ &= 1 - \frac{2K}{qm} \frac{t^2}{2!} + \frac{2K^2(2+q)}{q^2 m^2} \frac{t^4}{4!} - \frac{4K^3(q^2+2q+2)}{q^3 m^3} \frac{t^6}{6!} + \mathcal{O}(t^8) \end{aligned} \quad (27)$$

by the inverse Laplace transformation.

As we can see from the last equation, the short-time asymptotic of  $a(t)$  is characterized by the Gaussian behaviour and defined by inertial properties of the particle itself, whereas the long-time behaviour of the particle is independent of its mass (see equation (24)), but it is defined by the characteristics of the surroundings,  $K$  and  $m$ .

Then the Mori coefficients for  $a(t)$  are

$$\begin{aligned} \Omega_1^2 &= \omega^{(2)} = \frac{2K}{qm}, & \Omega_2^2 &= \frac{\omega^{(4)}}{\omega^{(2)}} - \omega^{(2)} = \frac{K}{m}, \\ \Omega_3^2 &= \frac{1}{\Omega_2^2} \left[ \frac{\omega^{(6)}}{\omega^{(4)}} - \left( \frac{\omega^{(4)}}{\omega^{(2)}} \right)^2 \right] = \frac{K}{m}. \end{aligned} \quad (28)$$

As a result, we find the first three points of the generalized non-Markovity parameter  $\varepsilon_n$  [6]:

$$\varepsilon_0 = \Omega_1^2 \tau_0^2 = \frac{1}{2}q, \quad \varepsilon_1 = \frac{\Omega_2^2}{\Omega_1^4 \tau_0^2} = 1, \quad \varepsilon_2 = \frac{\Omega_1^4 \Omega_3^2 \tau_0^2}{\Omega_2^4} = 1. \quad (29)$$

So, the non-Markovity parameter  $\varepsilon_0$  is proportional to the mass ratio  $q$ . This helps us to understand the reason for the observed Markovization at  $q \gg 1$  [37]. The non-Markovity parameter is also  $\varepsilon_0 \gg 1$ , which is true in Markovian case, when the time scale of memory is much smaller than the relaxation time of the velocity autocorrelation function. Another result concerning the unit values of the parameters  $\varepsilon_1$  and  $\varepsilon_2$  is also of interest. In particular, the value of  $\varepsilon_1 = 1$  means the equality of  $\tau_2$  and  $\tau_1$ , which has recently been discovered for Lennard-Jones fluids [6]. Furthermore, in case of  $M = 2m$  (i.e.  $q = 2$ ), the relaxation time scales of  $a(t)$ ,  $M_1(t)$  and  $M_2(t)$  are absolutely identical, and functions themselves may have the same time dependence, which can be exactly defined by the Bessel function of the first order (see the second equation of the system (25)).

The main results of this work can be summarized as follows:

- (i) Detailed numerical estimation of memory effects in diffusion processes of Lennard-Jones fluids has been executed for a wide phase range. For this purpose, the first three points of the generalized non-Markovity parameter have also been obtained for the velocity autocorrelation function in the case of the Rubin model.
- (ii) The results related to a unit value of  $\varepsilon_1$  for the Rubin model are evidence of equality of time scales of the first- and second-order memory functions (that was earlier found for the case of Lennard-Jones fluids [6]). It is important that this result is obtained here exactly from the primary model conditions.
- (iii) Linear interrelation between diffusion coefficient and non-Markovity parameter for the case of Lennard-Jones fluids has been found.

- (iv) The attempt to find interrelation between a measure of memory and the thermodynamic quantity, entropy, has also been made in this work. The obtained relation is evidence that ordering (disordering) processes in a system and non-Markovity of particle movements have immediate link.

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