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Mass-transfer in the Dusty Plasma as a Strongly Coupled Dissipative System: Simulations and Experiments

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1. Introduction

The problems associated with the mass-transfer processes in dissipative systems of interacting particles are of great interest in various fields of science (plasma physics, medical industry, physics and chemistry of polymers, etc.) (Frenkel, 1946; Cummins & Pike, 1974; Balescu, 1975; March & Tosi, 1995; Ovchinnikov et al., 1989; Dodd et al., 1982; Thomas & Morfill, 1996; Fortov et al., 1996; Fortov et al., 1999). Nevertheless, the hydrodynamic approaches can successfully describe these processes only in the case of the short-range interactions between particles. The main problem involved in studies of non-ideal systems is associated with the absence of an analytical theory of liquid. To predict the transport properties of non-ideal systems, the various empirical approaches and the computer simulations of dynamics of the particles with the different models for potentials of their interaction are used (Frenkel, 1946; Cummins & Pike, 1974; Balescu, 1975; March & Tosi, 1995; Ovchinnikov et al., 1989). The simulations of transport processes are commonly performed by methods of molecular dynamics, which are based on solving of reversible motion equations of particles, or Langevin equations taking into account the irreversibility of the processes under study.

The diffusion is the basic mass-transfer process, which defines the losses of energy (dissipation) in the system of particles and its dynamic features (such as the phase state, the conditions of propagation of waves and the formation of instabilities). When the deviations of the system from the statistical equilibrium are small, the kinetic coefficients of linear dissipative processes (constants of diffusion, viscosity, thermal conductivity etc.) can be found from Green-Kubo formulas that were established with the help of the theory of Markovian stochastic processes under an assumption of the linear reaction of the statistical system on its small perturbations. These formulas are the important results of the statistical theory of irreversible processes. According to these formulas, the diffusion coefficient D can be found from the following relationship:

$$D = \int_0^\infty \langle V(0)V(t)\rangle dt / m \,. \tag{1}$$

Here $<V(0)V(t)>$ is the velocity autocorrelation function (VAF) of grains, t is the time, and m is the dimension of the system. The diffusion coefficient can be also obtained from the analysis of a thermal transfer of the grains through the unit area of the medium:

$$D = \lim_{t\to\infty} <(\Delta l)^2>/(2mt), \tag{2}$$

where $\Delta l = \Delta l(t)$ is the displacement of an isolated particle from its initial position during the time t. In both equations (1), (2), the brackets < > denote the ensemble and time averaging (the averaging over all time intervals with the duration t). As the relationships (1)-(2) were obtained without any assumptions on a nature of a thermal motion, they are valid for gases as well as for liquids and solids in the case of the small deviations of the system from its steady state condition. In the general case of non-ideal fluids, the analytical solutions of Eqs.(1)-(2) are unavailable wich makes impossible to find the diffusion coefficient. The simple solution, $D \equiv D_o = T/(\nu_{fr}M)$, known as the Einstein relationship, exists only for the non-interacting ("brownian") particles; here M and T are the mass and the temperature of a grain, respectively, and v_{fr} is the friction coefficient.

Due to the existing level of experimental physics, it is necessary to go out of the bounds of diffusion approximation, and modern methods of numerical simulation (based on the theory of stochastic processes) allow one to make it. A description within the macroscopic kinetics may be insufficient for the analysis of mass-transfer processes on physically small time intervals. A study of the mass-transfer processes on short observation times is especially important for investigation of fast processes (e.g. the propagation of shock waves and impulse actions, or progression of front of chemical transformations in condensed matter (Ovchinnikov et al., 1989; Dodd et al., 1982)), and also for the analysis of transport properties of strongly dissipative media (such as colloidal solutions, plasma of combustion products, nuclear-induced high-pressure dusty plasma (Cummins & Pike, 1974; Fortov et al., 1996; Fortov et al., 1999)), where the long-term experiments should be carried out to measure the diffusion coefficients correctly.

2. Mass-transfer processes in non-ideal media

Consider the particle motion in a homogeneous dissipative medium. One can find a displacement of j-th particle in this medium along one coordinate, $x_j = x_j(t)$, under an action of some potential F and random F_{ran} forces from the Langevin equation

$$M\frac{d^2x_j}{dt^2} = -Mv_{fr}\frac{dx_j}{dt} + F + F_{ran}\,. \tag{3}$$

In a statistical equilibrium of system of particles ($M<(dx_j/dt)^2> = <MV_x(t)^2> \equiv T$) the mean value of the random force is zero, $<F_{ran}(t)> = 0$, and its autocorrelation function $<F_{ran}(0)F_{ran}(t)>= 2B\delta(t)$ corresponds to the delta-correlated Gaussian process, where $\delta(t)$ is the delta-function, and $B = T\nu_{fr}M$ (due to the fluctuation-dissipation theorem). Under these assumptions the Eq.(1) describes the Markovian stochastic process.

To analyze a dependence of mass-transfer on the time t, we introduce the following functions:

$$D_{G\text{-}K}(t) = \int_0^t \langle V_x(0)V_x(t)\rangle\, dt \tag{4a}$$

$$D_{msd}(t) = <(x_j)^2>/(2t) \tag{4b}$$

where $V_x(t)=dx_j/dt$ is the velocity of a j-th particle. With the small deviations of the system from the equilibrium state, both functions ($D_{G\text{-}K}(t)$ and $D_{msd}(t)$) with $t \to \infty$ should tend to the same constant $D = \lim_{t\to\infty} D(t)$, which corresponds to the standard definition of diffusion coefficient.

Neglecting the interparticle interaction ($F = 0$: the case of "brownian" particles), one can find the VAF, using the formal solution of Eq.(3) under assumption of $<F_{ran}(t)\ V_x(0)> = 0$ (Cummins & Pike, 1974):

$$\langle V_x(0)V_x(t)\rangle = \frac{T}{M}\exp\left(-\nu_{fr}t\right) \tag{5}$$

Then the mass-transfer evolution function $D_{G\text{-}K}(t)$, Eq.(4a), may be written as

$$D_{G-K}(t) = D_0\left(1-\exp\left(-\nu_{fr}t\right)\right) \tag{6a}$$

To find the mean-square displacement of a j-th particle, one should multiply Eq.(3) by x_j. Then, if there is no correlation between the slow particle motion and the "fast" stochastic impact ($<F_{ran}x_j>=0$), the simultaneous solution of Eqs.(3), (4b) in a homogeneous medium ($M<(dx_j/dt)^2> \equiv T$, $<(\Delta l)^2> = m<x_j^2>$) can be presented as (Ovchinnikov et al., 1989)

$$D_{msd}(t)/D_o = 1-\left(1-\exp\left(-\nu_{fr}t\right)\right)/\nu_{fr}t \tag{6b}$$

Thus, for the "brownian" case, when $t \to \infty$ and $v_{fr}t \gg 1$, we have $D_{G\text{-}K}(t) = D_{msd}(t) \to D_o$, and on small time intervals ($v_{fr}t \ll 1$) the motion of particles has a ballistic character: $<x^2>\equiv<x_j^2>\approx T\ t^2/M$ and $D_{msd}(t) = <x^2>/(2t) \propto t$.

The analytical solution of Eq. (3) may be also obtained for an ideal crystal under assumption that the restoring force $F = -M\omega_c^2 x_j$ acting on particles in lattice sites can be described by the single characteristic frequency ω_c (the case of harmonic oscillator). In this case we will have

$$M\frac{d^2x_j}{dt^2} = -Mv_{fr}\frac{dx_j}{dt} - M\omega_c^2 x_j + F_{ran} \tag{7}$$

After multiplying both parts of this equation by $x = x_j$, rearranging and averaging, taking into account that $\langle F_{ran}x\rangle = 0$ and $M<(dx/dt)^2> = 2<V_x(t)^2> \equiv T$), we will obtain (Vaulina et al., 2005b):

$$M\frac{d^2\langle x^2\rangle}{dt^2} = -Mv_{fr}\frac{d\langle x^2\rangle}{dt} - 2M\omega_c^2\langle x^2\rangle + 2T \tag{8}$$

Then the simultaneous solution of Eq.(8) and Eq. (4b) can be written as

$$\frac{D_{msd}(t)}{D_o} = \frac{1-\exp(-\nu_{fr}t/2)\left(\cosh(\nu_{fr}t\psi)+\sinh(\nu_{fr}t\psi)/\{2\psi\}\right)}{2\xi_c^2\nu_{fr}t} \tag{9}$$

where $\psi=(1-8\xi_c^2)^{1/2}/2$, and $\xi_c = \omega_c/\nu_{fr}$. In the case of $(1-8\xi_c^2) < 0$, the ψ value is imaginary: $\psi = i\psi^*$, where $\psi^* = (8\xi_c^2-1)^{1/2}/2$. In this case, $\sinh(i\psi^*\nu_{fr}t)=i\sin(\psi^*\nu_{fr}t)$, $\cosh(i\psi^*\nu_{fr}t)=\cos(\psi^*\nu_{fr}t)$,

and the expression for $D_{msd}(t)$ function will include the trigonometric functions instead of the hyperbolic functions.

To define VAF, $\langle V_x(0)V_x(t)\rangle \equiv \langle V_x(t_o)V_x(t_o+\tau)\rangle$, we will use the following designations: $V_x(t_o) \equiv V_0$, $V_x(t_o+\tau) \equiv V$, $X(t_o) = x$, $X(t_o+\tau) \equiv x + \Delta x$. The Eq.(7) can be presented as two expressions at two various instants of time ($t = t_o$ and $t = t_o+\tau$). Then, we can multiply the first of them by V, and the second by V_o, respectively. The sum of these two expressions, averaged on the particle's ensemble for all time intervals with the duration $t = \tau$ (taking into account, that $<x\Delta x>=0$, $\langle F_{ran}(t_o)V(t_o+\tau)\rangle = \nu_{fr} M\langle V(t_o)V(t_o+\tau)\rangle$, $\langle F_{ran}(t_o+\tau)V(t_o)\rangle = 0$ (Cummins & Pike, 1974; Ovchinnikov et al., 1989)), can be written as

$$\frac{d\langle V_o V\rangle}{dt} = -v_{fr}\langle V_o V\rangle - \omega_c{}^2 \frac{d\langle x^2\rangle}{dt} \tag{10}$$

The solution of this equation is

$$<V_o V>= \frac{1}{2}\frac{d^2\langle x^2\rangle}{dt^2}, \tag{11}$$

and the Eq.(10) may be rewritten as

$$\frac{d^2\langle V_o V\rangle}{dt^2} = -v_{fr}\frac{d\langle V_o V\rangle}{dt} - 2\omega_c{}^2\langle V_o V\rangle . \tag{12}$$

Thus, in this case of harmonic oscillator, we will have for VAF

$$\langle V_x(0)V_x(t)\rangle = \frac{T}{M}\exp\left(-\nu_{fr}t/2\right)\left(\cosh(\nu_{fr}t\psi) - \sinh(\nu_{fr}t\psi)/\{2\psi\}\right) \tag{13}$$

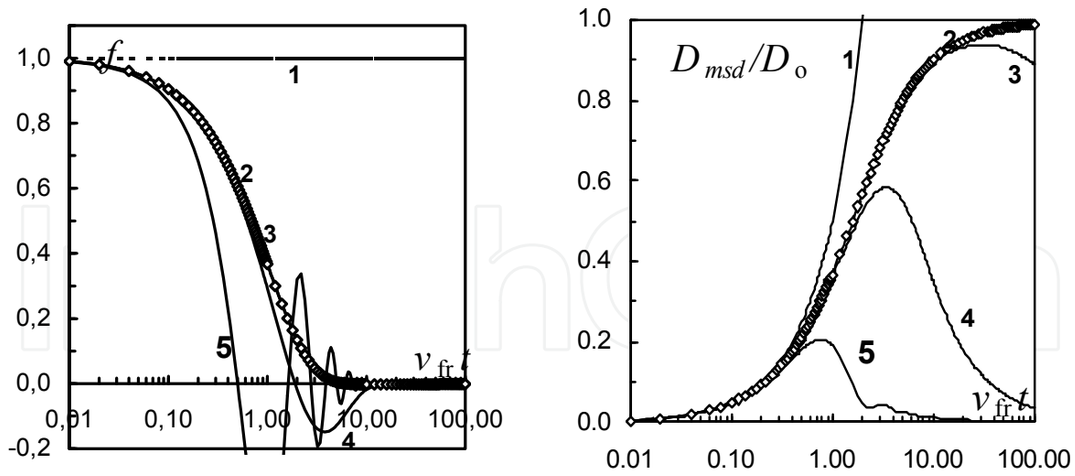
and the simultaneous solution of Eq.(13) and Eq.(4a) can be written as

$$\frac{D_{G-K}(t)}{D_0} = \frac{\exp\left(-\nu_{fr}t/2\right)}{\psi}\cdot\sinh\left(\nu_{fr}t\psi\right). \tag{14}$$

When ψ is imaginary, the both expressions for VAF and $D_{msd}(t)$ functions (Eqs. (13), (14)) will include the trigonometric functions instead of the hyperbolic ones (see above).

The normalized VAF, $f(t) = M <V_x(0)V_x(t)>/T$, and mass-transfer evolution functions ($D_{G-K}(t)/D_o$, $D_{msd}(t)/D_o$) for various values of ξ_c are presented in Fig. 1, where the time is given in units of inverse friction coefficient (v_{fr}^{-1}). It is easy to see that for short observation times a particle in a lattice site also has the ballistic character of motion ($<x^2> \approx T t^2/M$, $D_{msd}(t) = <x^2>/(2t) \propto t$). With the increasing time ($v_{fr}t \gg 1$) both evolution functions tend to zero: $D_{G-K}(t) = D_{msd}(t) \to 0$, because for the harmonic oscillator the mean-square declination $<(\Delta l)^2>$ is constant: $<(\Delta l)^2> = mT/(M\omega_c^2)$.

For liquid media, the exact analytic expression for $<V_x(0)V_x(t)>$, $D_{G-K}(t)$ and $D_{msd}(t)$ can't be obtained. Nevertheless, we should note some features referring to the relations between the mentioned functions, both in the case of "brownian" particles (see Eqs. (5)-(6b)) and in the case of harmonic oscillator (see Eq. (13)), which may take place for liquids:



(a) (b)

Fig. 1. Functions $f(tv_{fr})$ (**a**) and $D_{msd}(tv_{fr})/D_o$ (**b**) for : **1** – ballistic mode ($f(t)$=1, $D_{msd}(t) \propto t$); **2** – "brownian" case (Eqs.(5), (6b)); and for harmonic oscillator (Eqs.(13), (9)) with the different ξ_c: **3**– 0.033; **4** – 0.38; **5** – 2.

$$D_{G\text{-}K}(t) = \frac{d\{tD_{msd}(t)\}}{dt} \equiv \frac{1}{2}\frac{d\left\langle x^2 \right\rangle}{dt}, \tag{15a}$$

$$<V_x(0)V_x(t)>= \frac{d^2\{tD_{msd}(t)\}}{dt^2} \equiv \frac{1}{2}\frac{d^2\left\langle x^2 \right\rangle}{dt^2}. \tag{15b}$$

The mean-square displacement evolution $D_{msd}(t)$ was studied numerically in (Vaulina et al., 2005b; Vaulina & Dranzhevski, 2006; Vaulina & Vladimirov, 2002) for non-ideal systems with a screened Coulomb pair interaction potential (of Yukawa type):

$$U = (eZ)^2\exp(-r/\lambda)/r\,. \tag{16}$$

Here r is a distance between two particles with a charge eZ, where e is an electron charge, λ is a screening length, $\kappa = l_p/\lambda$, and l_p is the mean interparticle distance, which is equal the inverse square root from the surface density of particles for two-dimensional (2d-) systems, and it is the inverse cubic root of their bulk concentration in three-dimensional (3d-) case. As a result of numerical simulation, the characteristic frequencies for the body-centered cubic (*bcc*-) lattice ($\omega_c = \omega_{bcc} \approx 2eZ\exp(-\kappa/2)[(1+\kappa+\kappa^2/2)/(l_p^3M\pi)]^{1/2}$) and for hexagonal lattice ($\omega_c = \omega_h \approx 1.16\omega_{bcc}$) were obtained (Vaulina et al., 2005b; Vaulina & Dranzhevski, 2006; Vaulina & Vladimirov, 2002). It was also shown that these frequencies are responsible for the mean time t_a of "settled life" of particles in liquid-like systems and their values define the evolution of mass-transfer for the observation times $t < t_a \approx 2/\omega_c$. Taking into account Eqs.(15a)-(15b), it is easy to assume that the behavior of VAF and $D_{G\text{-}K}(t)$ for the mentioned observation times in liquid-like Yukawa systems is also close to the behavior of these functions for harmonic oscillator .

The dynamics of 3d- systems of particles with the various types of pair isotropic potentials was numerically investigated in (Vaulina et al., 2004). Those potentials represented different

combinations of power-law and exponential functions, commonly used for simulation of repulsion in kinetics of interacting particles (Ovchinnikov et al., 1989):

$$U = U_c [b_1 \exp(-\kappa_1 r/l_p) + b_2 (l_p/r)^n \exp(-\kappa_2 r/l_p)]. \quad (17)$$

Here $b_{1(2)}$, $\kappa_{1(2)} = l_p/\lambda_{1(2)}$ and n are variable parameters, and $U_c = (eZ)^2/r$ is the Coulomb potential. In the context of investigation of dusty plasma properties, the screened Coulomb potential (16) (b_1=1, b_2=0, $\kappa_1 = l_p/\lambda$) is of particular interest. But it should be noted that the simple model (16) agrees with numerical and experimental results in a complex plasma only for short distances $r < \lambda$ between two isolated macro-particles in a plasma (Konopka et al., 1997; Daugherty et al., 1992; Allen, 1992; Montgomery et al., 1968). With increasing distance, the effect of the screening weakens, and the asymptotic character of the potential U for large distances $r >> \lambda$ can follow the power-law dependence: $U \propto r^{-2}$ (Allen, 1992) or $U \propto r^{-3}$ (Montgomery et al., 1968); thus, the parameters of the potentials (17) will be $\kappa_1 = l_p/\lambda$; κ_2 =0; n =1-2; $b_1 >> b_2$, respectively.

It was noticed that the mass-transfer processes and spatial correlation of macroparticles in these 3d- systems are defined by the ratio of the second derivative U'' of a pair potential $U(r)$ in the point of the mean interparticle distance $r = l_p$ to the grains' temperature T, if the following empirical condition is met (Vaulina et al., 2004):

$$2\pi > | U'(l_p) | l_p / | U(l_p) | > 1. \quad (18)$$

In this case, the spatial correlation of particles didn't depend on the friction (ν_{fr}) and was defined by the value of effective coupling parameter $\Gamma^* = M l_p^2 U''/(2T)$ in the range between Γ^*~10 and the point of crystallization of the system ($\Gamma^* \sim 100$), where for all considered cases the formation of *bcc*- structure was observed with the characteristic oscillation frequency of the grains (Vaulina et al., 2004):

$$\omega_c^2 = \omega_{bcc}^2 \equiv 2 | U''(l_p) | / (\pi M). \quad (19a)$$

It is to expect that the characteristic oscillation frequency in the hexagonal lattice for the grains, interacting with the potentials (17), may be written similarly to the frequency found for the quasi-2d- Yukawa systems (Vaulina et al., 2005b; Vaulina & Dranzhevski, 2006):

$$\omega_c^2 = \omega_h^2 \equiv 2.7 | U''(l_p) | / (\pi M). \quad (19b)$$

The behavior of VAFs and of the evolution functions, $D_{msd}(t)$, $D_{G\text{-}K}(t)$, are studied numerically in the next part of this paper for quasi-2d- and 3d- non-ideal systems with the different interaction potentials, which obey the Eqs.(17),(18).

3. Parameters of numerical simulation

The simulation was carried out by the Langevin molecular dynamics method based on the solution of the system of differential equations with the stochastic force F_{ran}, that takes into account processes leading to the established equilibrium (stationary) temperature T of macro-particles that characterizes kinetic energy of their random (thermal) motion. The simulation technique is detailed in Refs. (Vaulina et al., 2005b; Vaulina & Dranzhevski, 2006; Vaulina & Vladimirov, 2002; Vaulina et al., 2003). The considered system of N_p motion

equations (N_p is a number of grains) included also the forces of pair interparticle interaction $\boldsymbol{F}_{int}$ and external forces $\boldsymbol{F}_{ext}$:

$$M\frac{d^2\vec{l}_k}{dt^2} = \sum_j F_{int}(l)\Big|_{l=|\vec{l}_k-\vec{l}_j|}\frac{\vec{l}_k-\vec{l}_j}{|\vec{l}_k-\vec{l}_j|} + \vec{F}_{ext} - M\nu_{fr}\frac{d\vec{l}_k}{dt} + \vec{F}_{ran} \qquad (20)$$

Here $F_{int}(l) = -\frac{\partial U}{\partial l}$, and $l = |\vec{l}_k - \vec{l}_j|$ is the interparticle distance. To analyze the equilibrium characteristics in the systems of particles interacting with potentials (17), the motion equations (20) were solved with various values of effective parameters that are responsible for the mass transfer and phase state in dissipative non-ideal systems. These parameters were introduced by analogy with the parameters found in (Vaulina & Dranzhevski, 2006; Vaulina & Vladimirov, 2002; Vaulina et al., 2004), namely the effective coupling parameter

$$\Gamma^* = a_1 l_p^2 U''(l_p)/(2T), \qquad (21)$$

and the scaling parameter

$$\xi = \omega^*/v_{fr}, \text{ where } \omega^* = |\, a_2 U''(l_p)\,|^{1/2}(2\pi M)^{-1/2}. \qquad (22)$$

Here $a_1 = a_2 \equiv 1$ for 3d- systems, and $a_1 = 1.5$, $a_2 = 2$ for quasi-2d- case. The calculations were carried out for a uniform 3d- system and for a quasi-2d- system simulating an extensive dusty layer. The scaling parameter was varied from $\xi \approx 0.04$ to $\xi \approx 3.6$ in the range typical for the laboratory dusty plasma in gas discharges; thus, the values of Z were varied from 500 to 50000, the particle mass, M, was 10^{-11} - 10^{-8} g, and the l_p values were ~100-1000 μm. The Γ^* value was varied from 10 to 120.

In the 3d- case the external forces were absent ($|\boldsymbol{F}_{ext}| \equiv 0$), and the periodical boundary conditions were used for all three directions $\boldsymbol{x}$, $\boldsymbol{y}$ and $\boldsymbol{z}$. The greater part of calculations was performed for 125 independent particles in a central calculated cell that was the cube with the characteristic size L. The length of the cell L (and the corresponding number of particles) was chosen in accordance with the condition of a correct simulation of the system's dynamics: $L >> l_p|U(l_p)|/\{|U'(l_p)|l_p - |U(l_p)|\}$, that satisfies the requirement of strong reducing of the pair potential at the characteristic distance L (Vaulina et al., 2003). So, for example, for Yukawa potential this conditions may be presented in the form $l_p/L << \kappa$ (Totsuji et al., 1996). The potential of interparticle interaction was cut off on the distance $L_{cut} \sim 4\, l_p$, which was defined from the condition of a weak disturbance of electrical neutrality of the system: $U'(L_{cut})\, L_{cut}^2 << (eZ)^2$. To prove that the results of calculation are independent of the number of particles and the cutoff distance L_{cut}, the additional test calculations were carried out for 512 independent particles with $L_{cut} = 7\, l_p$ and $\Gamma^* = 1.5$, 17.5, 25, 49 and 92. The disagreement between the results of these calculations was within the limits of the numerical error and didn't exceed ± (1-3)%.

In the quasi-2d- case, the simulation was carried out for the monolayer of grains with periodical boundary conditions in the directions $\boldsymbol{x}$ and $\boldsymbol{y}$. In $\boldsymbol{z}$ direction the gravitational force Mg, compensated by the linear electrical field $E_z = \beta z$ ($|\boldsymbol{F}_{ext}| \equiv F_{ext}^z = Mg - eZ\beta z$), was considered. Here β is the gradient of electrical field, and $F_{ext}^x = F_{ext}^y \equiv 0$. The number of independent particles in the central calculated cell was varied from 256 to 1024; accordingly,

the cutoff distance of potential was changed from $5l_p$ to $25l_p$. The value of the gradient β of electrical field E_z, confining the layer in z direction, was varied from ~10^{-2} V/cm^2 to ~100 V/cm^2, and for the simulated monolayers of grains the β value was in an agreement with the criterion of formation of mono-layer's dust structures proposed in (Vaulina et al., 2005a): $eZ\beta < 2\sum_{i=1}^{N_p} U'(l_i)/l_i$. Under this condition we have not detected any considerable dependence of particles' dynamics on the values of β and N_p in our simulations.

4. Results of the numerical simulation and their discussion

The evolution of mass-transfer processes, obtained in the numerical experiments for quasi-2d- and 3d- systems with various interaction potentials for different values of ξ and Γ^* is illustrated in Figs. 2-5 where the normalized VAFs, $f(v_{fr}t) = M\langle V(0)V(v_{fr}t)\rangle/(mT)$, and mass-transfer evolution functions ($D_{G\text{-}K}(v_{fr}t)/D_o$, $D_{msd}(v_{fr}t)/D_o$) are presented. In Figs. 2-3, the curves 1 are the solution of Langevin equation with neglecting of the interparticle interaction (see Eqs. (5)-(6 a,b)). It can be easily seen that in the presence of interparticle interactions, the behavior of $\langle V(0)V(t)\rangle$, $D_{msd}(t)$, $D_{G\text{-}K}(t)$ on short observation times ($v_{fr}\,t \ll 1$) corresponds to the motion typical for "brownian" particles. With time, the functions $D_{msd}(t)$, $D_{G\text{-}K}(t)$ reach their maximums D_{msd}^{max} and $D_{G\text{-}K}^{max}$. However, neither the relative magnitude D_{msd}^{max}/D_o, $D_{G\text{-}K}^{max}/D_o$ nor the position $t_{max}v_{fr}$ of these maximums depend on Γ^* and are defined by the value of the scaling parameter ξ for 3d- problem as well as for the simulated 2d- system. This feature was noticed earlier for the functions $D_{msd}(t)$ (Vaulina et al., 2005b; Vaulina & Dranzhevski, 2006; Vaulina & Vladimirov, 2002; Vaulina et al., 2004).

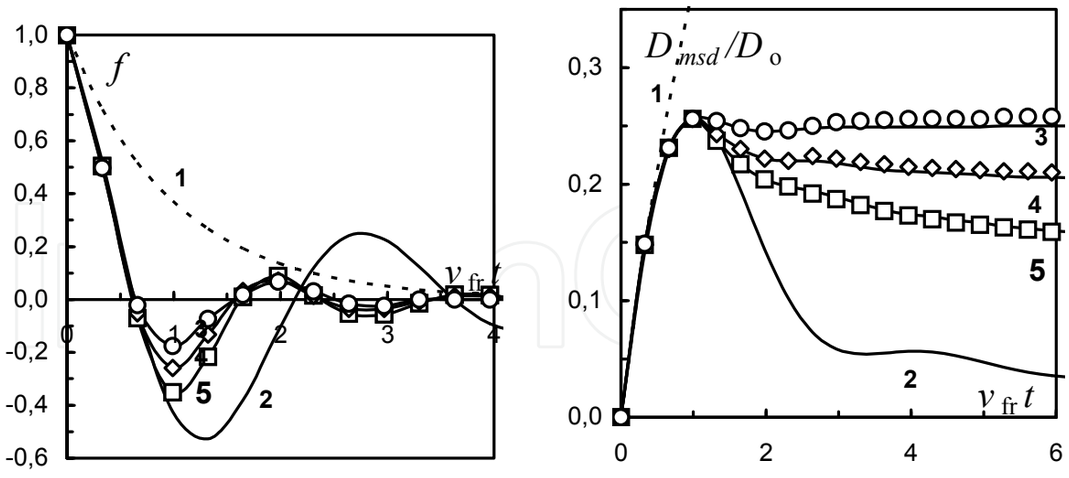


Fig. 2. Functions $f(tv_{fr})$ (**a**) and $D_{msd}(tv_{fr})/D_o$ (**b**) for: **1** – "brownian" case; **2** – harmonic oscillator with ξ_c =1.53. And the numerical results for quasi-2d- problem with ξ= 0.93 (ξ_c =1.53) and various Γ^*: **3** – 12; **4** – 27; **5** – 56; and for different potentials U: solid lines - $U/U_c = \exp(-4r/l_p)$; ○- $U/U_c = 0.1\exp(-2r/l_p) + \exp(-4r/l_p)$; ◇- $U/U_c = \exp(-4r/l_p) + 0.05\, l_p/r$; □- $U/U_c = 0.05(l_p/r)^3$.

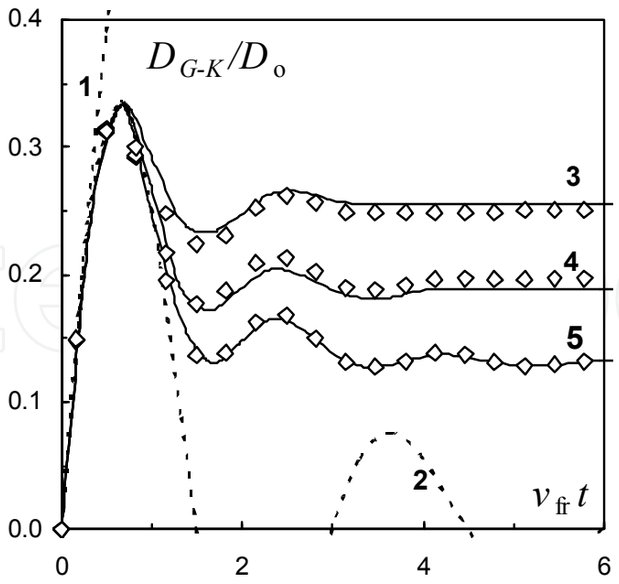


Fig. 3. Functions $D_{G\text{-}K}(tv_{fr})/D_o$ for: (**1**) - "brownian" case; (**2**) - for harmonic oscillator with ξ_c =1.53. And the numerical results for quasi-2d- problem (ξ = 0.93, ξ_c =1.53) with different Γ^*: **3** – 12; **4** – 27; **5** – 56. Fine lines - $D_{G\text{-}K}/D_o$ (Eq. (4a)), ◇- $D_{G\text{-}K}/D_o$ (Eq. (15a).

It is easy to see that the evolution of $<V(0)V(t)>$, $D_{msd}(t)$, $D_{G\text{-}K}(t)$ functions for the systems with the different pair potentials is defined by the particle temperature, T, the effective coupling parameter, Γ^*, the scaling parameter ξ (see Figs. 2-5) and also that the relation between $D_{msd}(t)$ and $D_{G\text{-}K}(t)$ is in accordance with Eq. (15a) (see Fig. 3).

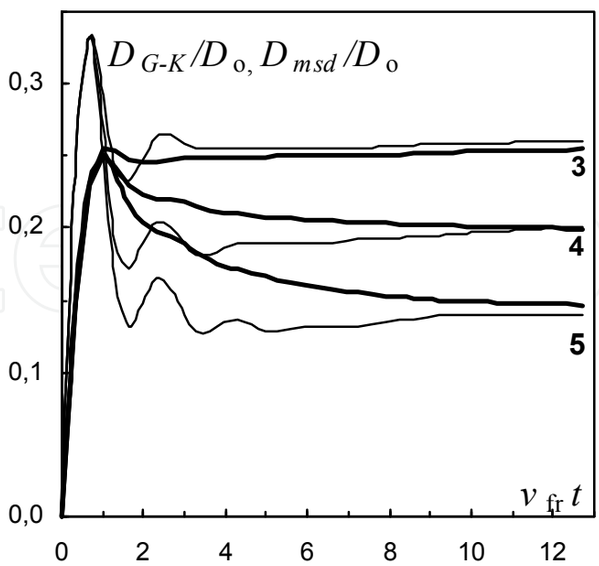


Fig. 4. Functions $D_{G\text{-}K}(tv_{fr})/D_o$ (fine lines, Eq. (4a)) and $D_{msd}(tv_{fr})//D_o$ (thick lines, Eq. (4b)) for quasi-2d- problem(ξ= 0.93, ξ_c =1.53) with different Γ^*: **3** – 12; **4** – 27; **5** – 56.

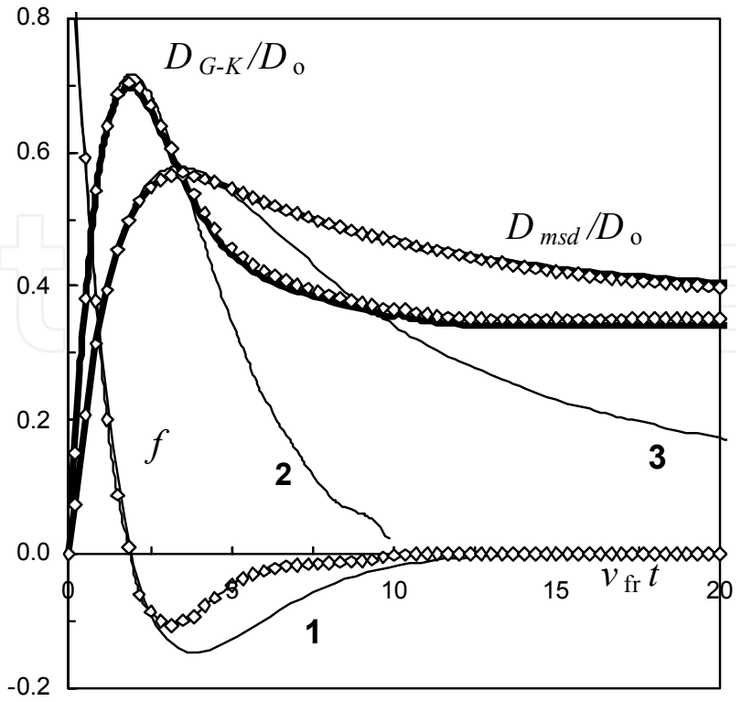


Fig. 5. Functions $f(tv_{fr})$ (**1**), $D_{G\text{-}K}(tv_{fr})/D_o$ (**2**) and $D_{msd}(tv_{fr})/D_o$ (**3**) for harmonic oscillator (fine lines) with ξ_c =0.19. And the numerical results for 3d- problem with ξ= 0.19 (ξ_c =0.38) and Γ^* = 27 for different potentials U: (thick lines) - U/U_c =exp(-2.4r/l_p);◇- U/U_c = exp(-4.8 r/l_p) + 0.05 l_p/r.

With $t \to \infty$, both $D_{msd}(t)$ and $D_{G\text{-}K}(t)$ functions tend to the same constant value D that corresponds to the diffusion coefficient. The normalized coefficients $D^*=D(v_{fr}+\omega^*)M/T$ vs. the Γ^* parameter for quasi-2d- systems with various pair potentials are shown in Fig. 6. It can be easily noticed that the D^* value for the systems under study is defined by the value of Γ^*. It was observed that the difference between the diffusion coefficients of weakly dissipative (ξ > 0.3) and weakly dispersive (ξ < 0.25) quasi-2d- structures with Γ^* between ~6 and ~97 is rather small; within the mentioned range of Γ^* the deviations of diffusion coefficients from their mean value don't exceed 7%. This difference increases noticeably with an increase of Γ^* > 100. These deviations were observed for quasi-2d- Yukawa systems as well as for 3d- systems with different types of pair potentials (Vaulina et al., 2005b; Vaulina & Dranzhevski, 2006; Vaulina & Vladimirov, 2002; Vaulina et al., 2004). Note that the obtained functions $D^*(\Gamma^*)$ have two critical points, one of which is a point of inflection (Γ^*~ 98-108) that, possibly, reflects a phase transition between the hexatic phase and the liquid. The second critical point (the point of a abrupt change of D) lies near Γ^*~ 153-165, where $D \to 0$, and the system under study is transforming into the solid with a perfect hexagonal lattice. The similar behavior of $D^*(\Gamma^*)$ was observed for quasi-2d Yukawa systems (Vaulina & Dranzhevski, 2006). The mean value of normalized diffusion coefficient, D^*, averaged for different values of Z, κ, v_{fr} and β) for the quasi-2d- systems with the different potentials is presented in Fig. 7, where the dependence $D^*(\Gamma^*)$ for 3d-structures (Vaulina & Vladimirov, 2002; Vaulina et al., 2004) is also shown.

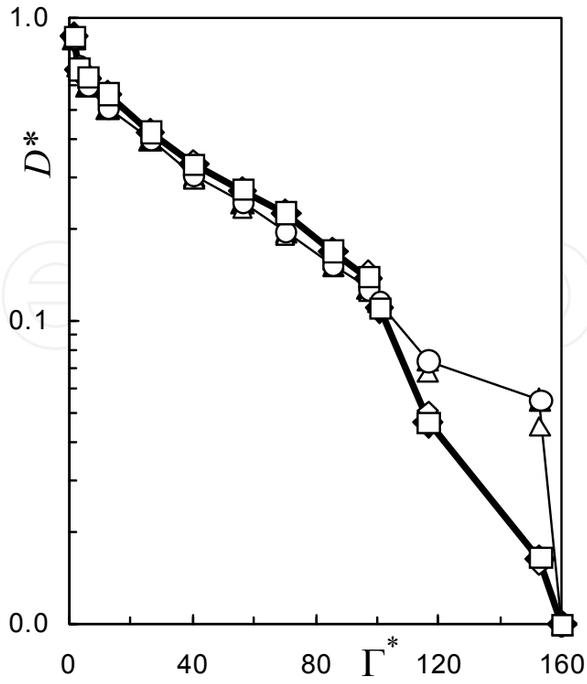


Fig. 6. Value of D^* vs. Γ^* for various potentials U: (black symbols) - $U/U_c = \exp(-4\ r/l_p) + 0.05\ l_p/r)$; (white symbols) - $U/U_c = 0.05(l_p/r)^3$; for different ξ: (◇, ◆) – 1.86; □ – 0.93, O – 0.23, (△, ▲) – 0.12. The solid lines are the averaged data of simulation for the quasy-2d- systems with $\xi < 0.25$ (thick line) and for $\xi > 0.3$ (fine line).

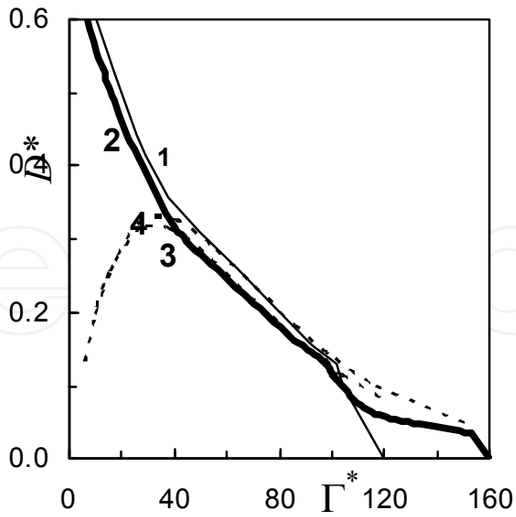


Fig. 7. Value of D^* vs. Γ^* for: **1 –** 3-d systems (see (Vaulina et al., 2004)); **2 –** quasi-2d- systems (averaged); **3 –** Eq.(23) for 2d- case with $\Gamma^*_c = 98$; **4 –** Eq.(23) for 3d- problem with $\Gamma^*_c = 102$.

The temperature dependence of the diffusion coefficient D of macroparticles in the 3d-systems with various types of potentials and for the quasi-2d- structures with the screened

Coulomb potential was found in (Vaulina & Dranzhevski, 2006; Vaulina & Vladimirov, 2002; Vaulina et al., 2004). There was shown that the diffusion coefficient for strongly coupled liquid-like systems can be presented as

$$D \approx \frac{T\Gamma^*}{12\pi(\xi+1)\nu_{fr}M}\exp\left(-3\frac{\Gamma^*}{\Gamma^*_c}\right), \tag{23}$$

where Γ^*_c is the crystallization point of the structure under study (Γ^*_c = 102 for the 3d-problem and Γ^*_c = 98 for the 2d- case). The approximation of the numerical results for obtained diffusion coefficients by Eq.(23) is shown in Fig. 7. The accuracy of this approximation, which for Γ^* >50 is just within 5%, decreases to 35% with Γ^* decreasing down to the value of $\Gamma^* \approx 30$ (Konopka et al., 1997; Daugherty et al., 1992; Allen, 1992). It should be noted that relation (23) is in accordance with the empirical "jumps" theory developed for molecular fluids that is based on the analogies between the liquid and the solid states of matter (Frenkel, 1946; March & Tosi, 1995); and it allows experimental determination of Γ^* in a strongly coupled system from measurements of the mean interparticle distance l_p, temperature T_p, and diffusion coefficient D without additional physical assumptions on the character of pair potential (Vaulina et al., 2003).

The comparison of evolution of mass-transfer processes in liquid-like 3d- and quasi-2d-systems with the behavior of analytical $D_{msd}(t)$, $<V_x(0)V_x(t)>$, $D_{G\text{-}K}(t)$ functions, obtained for harmonic oscillator, Eqs.(9),(13),(14), demonstrates a good agreement for observation times $\nu_{fr}\, t \le 1/\xi$ (see Figs. 2, 3, 5). Thus, in accordance with mentioned "jumps" theory the time of activation τ_o of "jumps" (the mean time of "settled life" of the particles) in the simulated systems practically doesn't depend on temperature and is defined by the oscillation frequency of the grains in "settled" condition: $\tau_o \approx 2/\omega_c$. The simulations also show that the system of interacting particles can be characterized by the constant values of transport coefficients only for intervals $t \gg \tau_o$, in contrast to the system of "brownian" particles, for which the evolution functions $D_{msd}(t)$ (or $D_{G\text{-}K}(t)$) tend to D_o for $t \gg \nu_{fr}^{-1}$.

The measurement of functions $<V(0)V(t)>$, $D_{msd}(t)$, $D_{G\text{-}K}(t)$ at the short observation times can be useful for the passive diagnostics of dust component in non-ideal plasma in the case of local statistical equilibrium of a dusty sub-system. As all the mentioned functions are connected by the relationships (15a)-(15b) and unambiguously depend on such parameters of grains as their temperature T, characteristic frequency ω_c and the friction coefficient ν_{fr}, it is possible to simultaneously determine all the mentioned parameters by measuring any of the functions $<V(0)V(t)>$, $D_{msd}(t)$ or $D_{G\text{-}K}(t)$ and using a procedure of best fitting of this chosen function by the respective analytical function for harmonic oscillator. Additionally, the information on T and ω_c allows one to estimate the value of the coupling parameter Γ^* of the system under study from the Eq. (21)-(22).

5. Mass transfer in the dusty plasma

The dusty plasma is an ionized gas containing micron-size charged grains (macroparticles) of solid matter (dust). This type of plasma is ubiquitous in nature (in space, in molecular dust clouds, in planetary atmospheres) and often appears in a number of technological processes (for example, fuel burning, an industrial processing of semiconductors etc) (Thoma et al., 2005; Morfill et al., 2003). The experiments with dusty plasma are carried out mostly in gas-discharges of various types. In gas-discharge plasma, micron-size grains

acquire a significant (negative) electric charge, and can form dust structures similar to a liquid or to a solid. Dependent on the experimental conditions, these structures can be close to the uniform three-dimensional (3d-) or to the non-isotropic quasi- two-dimensional (2d-) systems, which consist of several (usually from one up to ten) horizontal layers of macroparticles (Nunomura et al., 2006; Ratynskaia et al., 2006; Nosenko & Goree, 2004; Gavrikov et al., 2005; Nunomura et al., 2005). Owing to their size, the dust particles may be video-filmed (Ratynskaia et al., 2006; Nosenko & Goree, 2004; Gavrikov et al., 2005; Nunomura et al., 2005; Vaulina & Vladimirov, 2002; Vaulina et al., 2004). It makes the laboratory dusty plasma a good experimental model, which can be used to study various physical phenomena in systems of interacting particles, that attract widespread interest in the physics of non-ideal plasmas as well as in other areas such as plasma chemistry, physics of the atmosphere, medicine, physics of polymers etc.

A study of the kinetic (transport) coefficients (constants of diffusion, viscosity, thermal conductivity etc.) for dusty plasma is of great interest (Nunomura et al., 2006; Ratynskaia et al., 2006; Nosenko & Goree, 2004; Gavrikov et al., 2005; Nunomura et al., 2005). These constants are fundamental parameters that reflect the nature of interaction potentials and a phase state of the system. When the deviations of the system from the statistical equilibrium are small, the kinetic coefficients can be found from Green-Kubo formulas that were established with the help of the theory of Markovian stochastic processes under an assumption of the linear reaction of the statistical system on its small perturbations (March & Tosi, 1995; Ovchinnikov et al., 1989). The diffusion is the basic mass-transfer process, which defines the losses of energy (dissipation) in the system. The collisions of grains with the neutral particles of surrounding gas have a dramatic effect on the dissipation of dust energy in weakly ionized laboratory plasma.

The measurement of the $<V(0)V(t)>$-, $D_{msd}(t)$-, $D_{G\text{-}K}(t)$- functions at the short observation times can be used for the diagnostics of dust component in plasma. As all the mentioned functions are connected by the Eqs. (15a)-(15b) and uniquely determined by the parameters of grains (by their temperature T, characteristic frequency ω_c and the friction coefficient ν_{fr}), it is possible to simultaneously determine all these parameters using the best fitting of any from the measured functions ($<V(0)V(t)>$, $D_{msd}(t)$, $D_{G\text{-}K}(t)$) by the respective analytical function for harmonic oscillator. The information on T and ω_c allows one to estimate the value of the effective coupling parameter Γ^* and the scaling parameter ξ, which determine the dynamics of grains in the system under study.

Nevertheless we should note that to apply the results of simulation of particle motions by LMDM for analysis of dynamics of grains in plasma and to prove the validity of measurements of diffusion constants with the help of Green-Kubo formula (4a), one needs to examine, whether the considered Langevin model is valid under experimental conditions (i.e. to prove the validity of the Markovian approach of the condition of local equilibrium of dust system and of the assumption of the linear reaction of this system on its perturbations). The main objections to the use of the Langevin model in a treatment of the results of real experiments are the possible influence of boundary conditions, external fields and strong interparticle interactions on the migration of particles and their energy exchange with the surrounding medium (thermostat) in real experiments (Ovchinnikov et al., 1989). A lot of special questions arise in an interpretation of dusty plasma experiments. These are the influence of the openness of dusty plasma system and of the irregular distribution of grains' stochastic energy on the degrees of freedom, including the question on a capability of the use of this energy as the main thermodynamical characteristic that describes the kinetic

temperature of the dusty component and characterizes the exchange of its energy with a thermostat.

Thus, to verify a validity of the Langevin model, we should prove the diffusive character of the grains' migration (with $t \to \infty$: $\langle x^2 \rangle \propto t$, $D = const$), and also check the relations (15a), (15b), that result from the simulation of the dynamics of particles using the mentioned Langevin equations. (In the case when these relations are valid, the Green-Kubo formula (see Eq.(4a) with $t \to \infty$: $D_{G\text{-}K}(t) = D_{msd}(t) \equiv D$) is automatically true.)

6. The description of experiments and their results

The experiments were carried out for mono-disperse grains (with material density $\rho_p \approx 1.5$ g cm^{-3}, radiuses $a_p \approx 2.75$ μm and $a_p \approx 6.37$ μm) in the near-electrode area of RF-discharge in argon. The pressure in the discharge was $P = 0.03 - 0.5$ Torr, and its power – $W \approx 2\text{-}30$ W. The simplified scheme of the experimental setup is shown in Fig. 8. To visualize the cloud, a flat beam of He-Ne laser (λ = 633 nm) was used. The laser illumination had two regimes: in the first, the defocused laser beam illuminated the whole dusty structure inside the trap; this allowed to determine its dimensions. The second regime was used for the detailed observation of the horizontal cross-section of the dusty cloud. In this case the laser beam represented so called "laser knife" with the width ~ 2.5 cm and the waist size ~ 200 μm. The positions of grains were registered with a high-speed CMOS video camera (frame rate $f_{vc} = 500$ s^{-1}). The video-recording was processed by the special software, which allowed identifying the positions of each particle in the field of view of the video-system. Then the pair correlation functions $g(l)$, the mean inter-grain distances, the velocity autocorrelation functions ($\langle V(\delta t)V(t)\rangle = (\langle V_x(\delta t)V_x(t)\rangle + \langle V_y(\delta t)V_y(t)\rangle)/2$), the mass-transfer functions ($D_{G\text{-}K}(t)$, $D_{msd}(t)$), and the diffusion coefficients D were obtained. The deviations of the measured parameters in two registered freedom degrees (x, y) were minor and didn't exceed ~ 0.5-3%: $\langle V_x((\delta t)V_x(t)\rangle \cong \langle V_y(\delta t)V_y(t)\rangle$, $\langle V_x^2\rangle \cong \langle V_y^2\rangle$, $\langle x^2\rangle \cong \langle y^2\rangle$, where $\delta t = f_{vc}^{-1}$; and the value of the average dust velocities $\langle V_x(t)\rangle \cong \langle V_y(t)\rangle \equiv 0$.

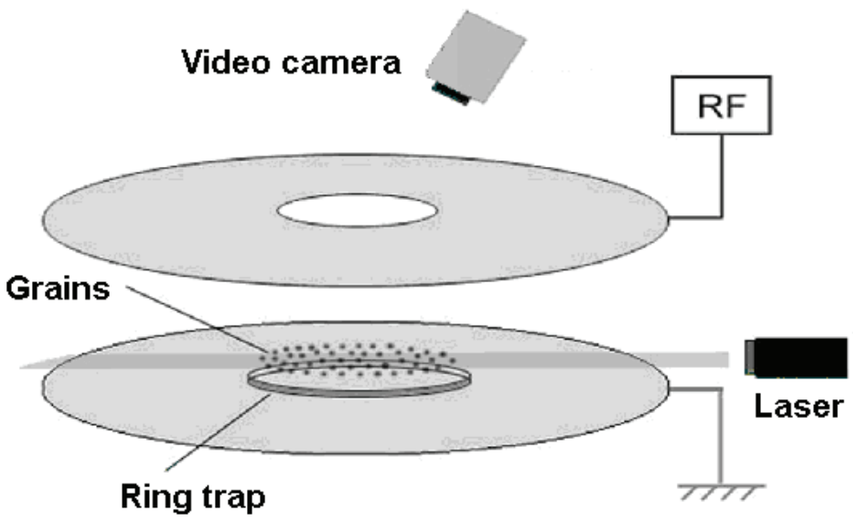


Fig. 8. The simplified scheme of experimental setup.

Under the experimental conditions, the dusty structures consisted of several (from 1 up to ~10) dusty layers, and the observed dusty structures were changing from the weakly correlated fluids to the dust crystals with the mean interparticle distance l_p from ~ 500 to ~1000 μm. The duration of one experiment under stable conditions was ~ 5-10 s. The experimental pair correlation functions $g(l)$ for the grains of different sizes, forming dusty mono-layers and multi-layer systems, are presented in Fig. 9.

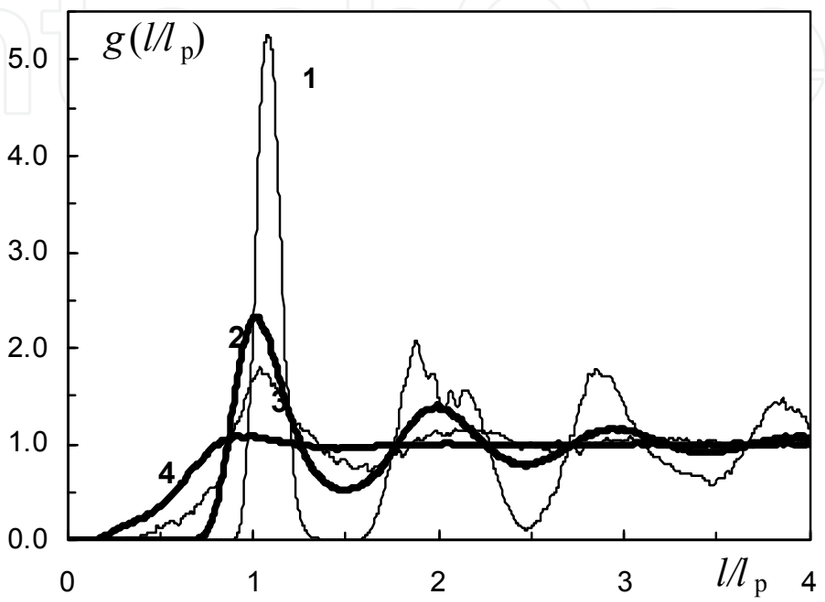


Fig. 9. The pair correlation function $g(l/l_p)$ for various experiments: **1** - a_p = 6.37 μm, mono-layer, P = 0.03 Torr; **2** - a_p = 2.755 μm, mono-layer, P = 0.35 Torr; **3** - a_p = 6.37 μm, multi-layer system, P = 0.11 Torr; **4** - a_p = 2.755 μm, multi-layer system, P = 0.04 Torr.

The magnitude of the maximum g_{max} of functions $g(l)$ and the mean interparticle distance l_p, which was obtained by analysis of the g_{max} position, are given in Table 1 for different experiments. The random errors of determination of the l_p and g_{max} values were less than 5% for all cases. Nevertheless, we should note that the magnitude of $g(l)$ peaks may be misrepresented because of: (i) limited number of grains in the field of view of the video camera; (ii) the probability of simultaneous registration of grains in the next layer in the case of the multi-layer structure.

The results of measurement of velocity autocorrelation functions, $\langle V(\delta t)V(t)\rangle$, and mass-transfer functions ($D(t)=D_{G\text{-}K}(t)$ and $D(t)=D_{msd}(t)$) for various experiments are presented in Fig. 10 and Fig. 11 where the normalized values are shown: $f(t)= \langle V(\delta t)V(t)\rangle / V_T^2$ and $D(t)/D_o$, where $V_T^2 = T/M$ is the mean square velocity of stochastic "thermal" motion of grains. The values of V_T^2 and ν_{fr} are presented in Table 1, and the methods of their determination are discussed in the next section.

For the most experiments, the motion of grains was diffusive: with the time increasing ($t\rightarrow \infty$), the mass-transfer functions tended to the same value: $D_{G\text{-}K}(t) \approx D_{msd}(t) \rightarrow D$, excluding the crystalline dusty structures, where with $t\rightarrow \infty$, the value of the mean square displacements of grain from its equilibrium position $\langle x^2\rangle \cong \langle y^2\rangle \rightarrow const$, and the D constant $\rightarrow 0$. The measured value of the diffusion coefficient D is given in Table 1. The

difference between the diffusion constants determined from the Green-Kubo formula (4a), $D = D_{G\text{-}K}(t\to\infty)$, and from the analysis of the mean square displacements, $D = D_{msd}(t\to\infty)$ (4b), didn't exceed 5%, that corresponds to the random error. The systematic errors in determination of D due to the finite duration of measurements were the same (≤ 5%). The time t_D of establishing the constant value of D (±5%) was from ~ 1 s to 5 s in the relation to the gas pressure P (with the increase of P, the t_D value was increasing also). With the low pressure P ($\xi_c \geq 1$), the $D_{msd}(t)$ function achieves its constant value, D, faster than the $D_{G\text{-}K}(t)$ function (see Fig. 11 a,c,d); otherwise with the higher pressure P ($\xi_c << 1$, the case of non-oscillating $D_{G\text{-}K}(t)$), the $D_{G\text{-}K}(t)$ value tends to the constant faster than $D_{msd}(t)$, that allows to measure the diffusion coefficient D from the Green-Kubo formula in shorter time (see Fig. 11b).

P, Torr	l_p, mm	g_{max}	V_T^2, mm²/s²	D, mm²/s	ν_{fr}, s^{-1}	ω_c, s^{-1}	Γ_{3d}/Γ_{2d}	Z_{min}
R = 2.75 μm, mono-layer								
0.11	1	2.75	0.803	0.0025	30.5	9.7	92/102	5605
0.19	0.84	2.55	0.720	0.002	50.8	9.8	72/80	4303
0.35	0.92	2.35	0.949	0.002	98	8.8	54/60	4478
0.5	0.75	2.7	1.468	0.00135	143	16.3	80/89	6119
R = 2.75 μm, multi-layer system								
0.04	1.1	1.1	26.3	0.81	11	13.2	6.3/7	8800
0.06	1	1.08	20.1	0.58	15.5	10.23	4.1/4.6	5912
0.1	0.57	1.095	20.7	0.3	34	22.44	6.2/6.9	5581
0.14	0.6	1.05	43.1	0.66	44	15.62	1.6/1.8	4195
R = 6.37 μm, mono-layer								
0.03	1	5.25	0.45	→ 0	3.5	13.0	293/326	26311
0.05	1	2.45	1.78	0.022	6	12.5	69/77	25396
0.08	0.88	2.75	1.23	0.01	8.4	13.5	90/100	22642
0.42	0.57	4.9	0.73	→ 0	44	27.3	262/291	23852
R = 6.37 μm, multi-layer system								
0.05	0.92	1.28	12.94	0.449	7	14	10/11	24964
0.07	0.82	1.74	4.51	0.095	8.2	16.4	31.5/35	24742
0.08	0.85	2	2.90	0.048	8.25	15	44/49	23883
0.11	0.88	1.66	4.97	0.12	11.8	13.1	21/23	21968

Table 1. Parameters of dusty component for various experiments

We emphasize, that under conditions of our experiments we have not observed the anomalous-, or super- diffusions that were investigated in a set of works (Ratynskaia et al., 2006; Wen-Tau & I, 1998). With increase of time t ($t\to\infty$), the both mass-transfer functions tended to the same value: $D_{G\text{-}K}(t) \approx D_{msd}(t) \to D$; i.e. the mean square displacements of particles were proportional to the time t for all presented experiments (see curves 1, 2 in Figs. 11 a,b,c,d).

The results of examination of relation between the VAFs, $\langle V(\delta t)V(t)\rangle$, mass-transfer functions ($D_{G\text{-}K}(t)$, $D_{msd}(t)$), and the mean-square displacement ($\langle \Delta l^2\rangle = \langle x^2\rangle + \langle y^2\rangle$) are presented in Figs. 10 - 11. In all the cases there was obtained a good agreement between the direct measurements of functions $\langle V(\delta t)V(t)\rangle$, $D_{G\text{-}K}(t)$ and those functions, calculated from the measurements of $\langle \Delta l^2\rangle$ using the Eqs. (15a)-(15b). Thus, we can conclude that the stochastic model, given by the system of Langevin equations, allows the correct description of the dust motion under experimental conditions.

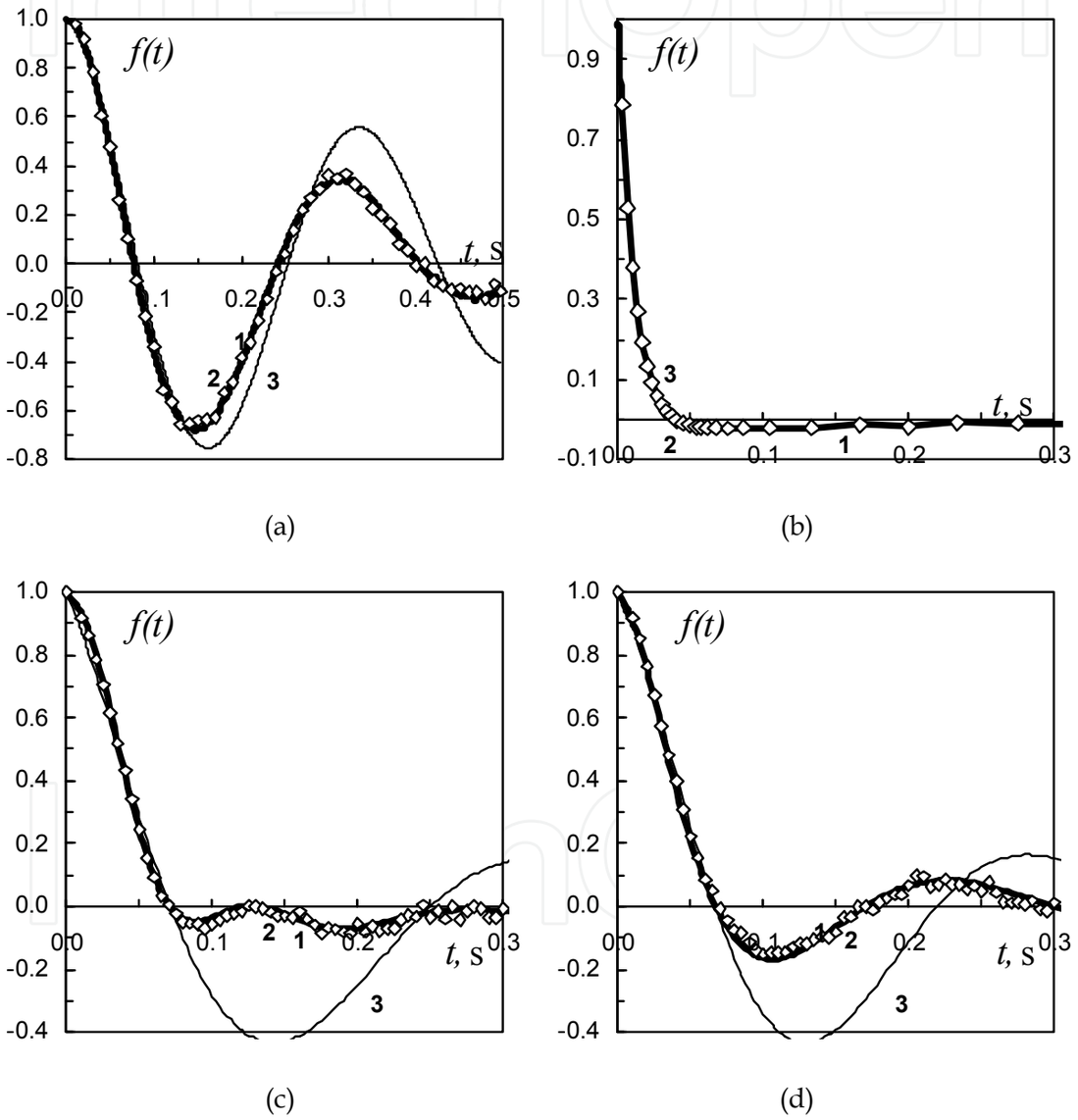


Fig. 10. The velocity autocorrelation function, $f(t)$ (curve **1**, line), and its value (curve **2**, ◇), obtained from the Eq.(15b), for various experiments: **(a)** - a_p = 6.37 μm, mono-layer, P = 0.03 Torr; **(b)** - a_p = 2.755 μm, mono-layer, P = 0.35 Torr; **(c)** - a_p = 6.37 μm, multi-layer system, P = 0.11 Torr; **(d)** - a_p = 2.755 μm, multi-layer system, P = 0.04 Torr.

Curve **3** is the $f(t)$ function for the harmonic oscillator, Eq.(13), with parameters indicated in Table 1.

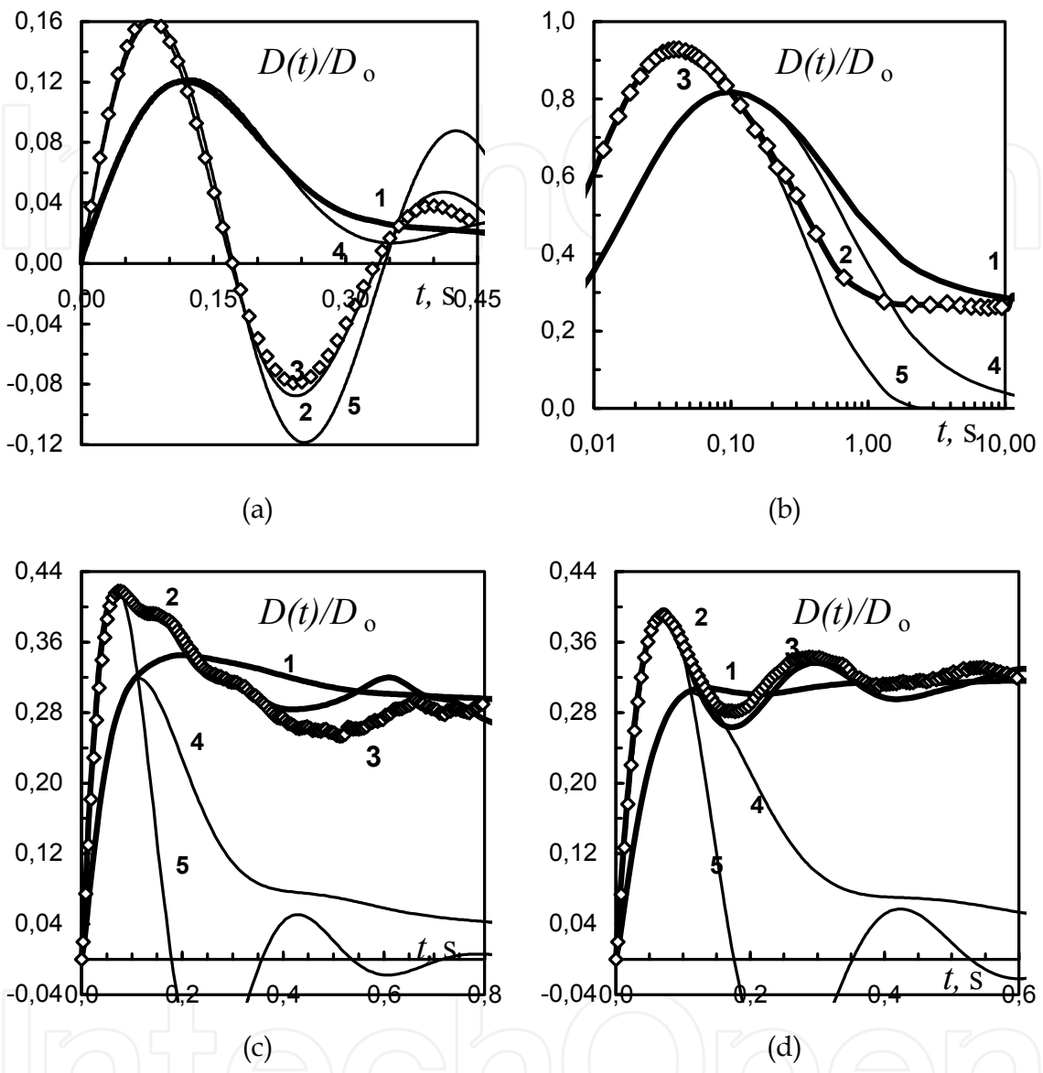


Fig. 11. The mass-transfer evolution functions $D(t)/D_o$: (curve **1**, line) – $D(t) \equiv D_{msd}(t)$; (curve **2**, line) - $D(t) \equiv D_{G\text{-}K}(t)$, Eq.(1b); (curve **3**, ◇) - $D(t) \equiv D_{G\text{-}K}(t)$, Eq.(15a), for various experiments: **(a)** - a_p= 6.37 μm, mono-layer, P=0.03 Torr; **(b)** - a_p= 2.755 μm, mono-layer, P = 0.35 Torr; **(c)** - a_p = 6.37 μm, multi-layer system, P=0.11 Torr; **(d)** - a_p= 2.755 μm, multi-layer system, P = 0.04 Torr.

Curve **4,** Eq.(9), and curve **5,** Eq.(14), are the corresponding functions for the harmonic oscillator with parameters indicated in Table 1.

7. The determination of parameters of dusty subsystem

The parameters of macroparticles (the mean-square velocity V_T^2 of their stochastic "thermal" motion, the characteristic frequency ω_c and the friction coefficient ν_{fr}) are presented in

Table 1. These parameters were obtained by the best fitting of the measured functions, $<V(\delta t)V(t)>$, $D_{msd}(t)$ and $D_{G\text{-}K}(t)$, and the corresponding analytical solutions, Eqs. (9, 13, 14), for the harmonic oscillator. The results of this procedure for various experiments are shown in Figs. 10-11. Note that the measured functions presented in these figures are relative, namely, they are the normalized values: $f(t)= <V(\delta t)V(t)>/V_T^2$ (see Fig. 10), and $D(t)/D_o$ (Fig. 11). The errors in determination of these relative functions for the same intervals of time, $t = (t' - t_o)$, were less than 2-3% for various initial time t_o which were chosen for processing experimental results. These random errors may be related to the fluctuations of dusty plasma parameters during experiments, and these errors were less than the errors in the determination of dusty plasma parameters (V_T^2 ,ν_{fr}, ω_c) by the best fitting of experimental data to the analytical solutions as discussed below.

The errors of the determined parameters were ~ 5-7% for V_T^2, less than 10% for the friction coefficient ν_{fr} , and less than 5% for ω_c. To optimize the procedure of fitting of experimental data, the initial values of ω_c and ν_{fr} were chosen in accordance with analytical approximations for the dependence of maximum D_{max} of function $D_{msd}(t)$ and the position of this maximum t_{max} on the parameter $\xi_c = \omega_c/\nu_{fr}$, that were proposed in (Vaulina & Dranzhevski, 2006; Vaulina et al., 2005b):

$$D_{max} \approx D_o/(1+ 2\xi_c). \tag{24}$$

$$t_{max}\nu_{fr} \approx 4\sqrt{2}\pi /(1+8\sqrt{2}\xi_c). \tag{25}$$

The accuracy of these approximations is about 5%. The Eq.(25) can be also used for choosing of the frame rate f_{vc} of video camera (registering the positions of grains) and the duration of measurement t_D that are necessary for the correct determination of dust parameters and diffusion coefficients D. So, the f_{vc} value should be much higher than $1/t_{max}$, and the duration t_D of measurement for determination of D should satisfy the condition $t_D >> t_{max}$. The preliminary estimation of values of ω_c and ν_{fr} under the experimental conditions may be performed using the existing theoretical models.

For all cases, the retrieved values of the mean-square velocity $V_T^2 = T/M$ were in accordance with the fitting of velocity distributions ($\varphi(V_x)$, $\varphi(V_y)$) by maxwellian functions (see Fig. 12). The difference between the values of V_T^2, obtained by two different methods, didn't exceed 5-7% and, in most cases, was within the limits of experimental error.

The retrieved values of ν_{fr} were in a good agreement with their theoretical estimations using the free-molecular approximation that under experimental conditions (for argon of the room temperature) may be presented as ν_{fr} [s^{-1}] $\approx$ 1144 P[Torr]/(a_p[μm]ρ_p[g/cm^3]) (Lifshitz & Pitaevskii, 1981; Raizer, 1991). The difference between the measured and theoretical values of ν_{fr} was less than 12% for all experiments.

For the direct examination of an accuracy of retrieved values for characteristic frequencies ω_c, we need an information on the form of pair potential U (see Eq.(19a,b)). Nevertheless, as the data on the dust parameters (ω_c ,V_T^2, l_p in Table 1) allows one to estimate the value of the effective coupling Γ^* and scaling ξ parameters (see Eqs.(19a,b, 21, 22)), we can compare the measured magnitudes of maximums, g_{max}, for pair correlation functions, and of diffusion coefficients, D, with existing numerical data. The values of Γ^*, obtained from solving Eqs.(19a,b,21), are presented in Table 1. For the case of mono-layer the value of $\Gamma^* = \Gamma_{2d}$ was determined in the 2-d approach ($a_o = 2.7$, $a_1 = 1.5$, $a_2 = 2$). For multi-layer systems the

estimations were performed for the two limiting cases: for 3-d case, $\Gamma^* = \Gamma_{3d}$ ($a_o = 2$, $a_1 = a_2 \equiv 1$), and for 2-d case, $\Gamma^* = \Gamma_{2d}$ (a_o=2.7, a_1 = 1.5, a_2 = 2).

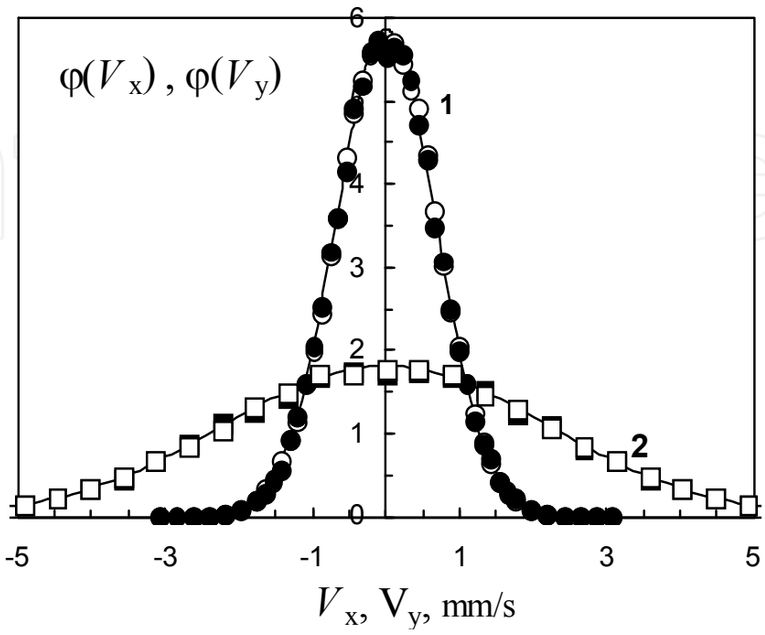
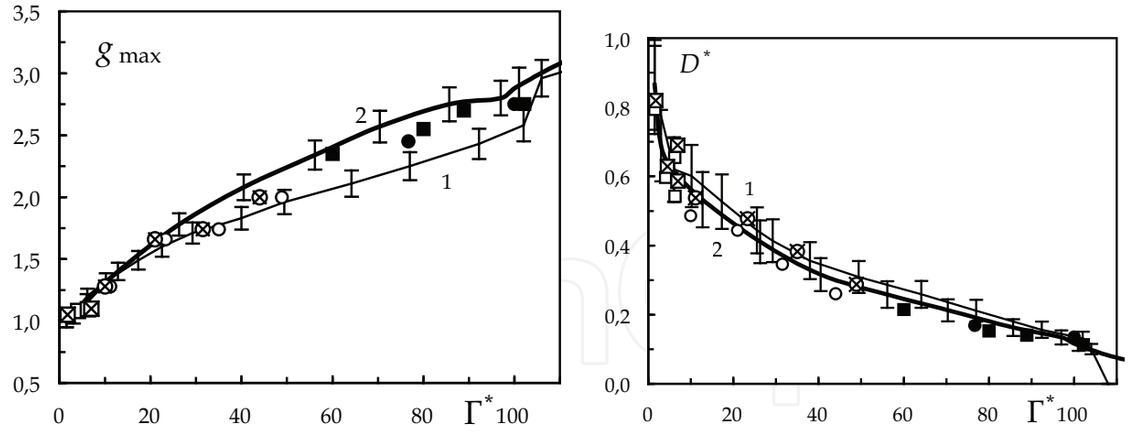


Fig. 12. The measured distribution of the velocities: φ(V_x) -(●; ■) and φ(V_y) -(○;□) - for the particles of the radius a_p = 6.37 μm and various experiments: (●;○) – mono-layer, P = 0.03 Torr; (■;□) - multi-layer system, P = 0.11 Torr. And their best-fit Maxwellian distributions with $V_T^2 = T/M$ equal to: **1** – 0.46 mm²/s²; **2** – 4.8 mm²/s².

The comparison of the experimental values of maximums, g_{max}, and of the normalized diffusion coefficient $D^* = v_{fr}\, D(1+\xi)M/T$ with the results, obtained via the numerical simulation for 3-d problem and mono-layer (Vaulina et al., 2004; Vaulina & Dranzhevski, 2006), is shown in Figs. 13a - 13b. (Here we note that the 2d- and 3d-systems were simulated here and in (Vaulina et al., 2004) for a wide range of pair isotropic potentials, and in (Vaulina & Dranzhevski, 2006) for the quasi-2d-Yakawa systems.) It is easy to see that the measured dependence $g_{max}(\Gamma^*)$ is in a good agreement with numerical data (see Fig.13a). The difference between them is in the limits of experimental (~5%, see Section 2) and numerical (~5%) errors in the determination of g_{max}. The values of errors are shown in Fig.13a as a 5% confidence interval. The measured dependence $D^*(\Gamma^*)$ is also in a good agreement with the numerical data for all considered experiments (Fig.13b). The deviations between the experimental and numerical values of D^* don't exceed the experimental (~10%, see Section 2), and numerical error, δ_{cal}, in the determination of D; the δ_{cal} value increases from 7% to 15% when Γ^* varies from ~ 100 to ~ 5.

The value of the minimum charge $Z = Z_{min}$, that a grain can acquire in plasma, may be obtained under the assumption that the surrounding plasma doesn't screen the interaction between grains. Thus, in the case of Coulomb interparticle interaction, we obtain $Z_{min} \cong \omega_c \{\pi M\, l_p^{\,3}/5.4\}^{1/2}$ (see Eq. (19a,b)). The values of Z_{min} are presented in Table 1. The error of this estimation of Z_{min} is determined by the experimental errors of ω_c and l_p and is about 13%.



(a) (b)

Fig. 13. Maximum g_{max} (**a**) of the $g(l)$ function and the normalized diffusion coefficient $D^*= D(v_{fr}+\omega^*)\, M/T$ (**b**) vs. Γ^* for: **1 –** 3d- systems; **2** – mono-layer (Vaulina et al., 2004; Vaulina & Dranzhevski, 2006). The symbols are the experimental results (Table 1) for the grains of radius a_p: (○; ●; ⊗)- 6.37 μm; (□; ■; ⊠) - 2.755 μm, that form the dusty mono-layer (●; ■), or multi-layer structure (○; □; ⊗; ⊠). For the multi-layer structure the values of ω^* and Γ^* were determined for two cases: (○; □) – 3d- system $\Gamma^* = \Gamma_{3d}$; (⊗; ⊠) – 2d- system, $\Gamma^* = \Gamma_{2d}$.

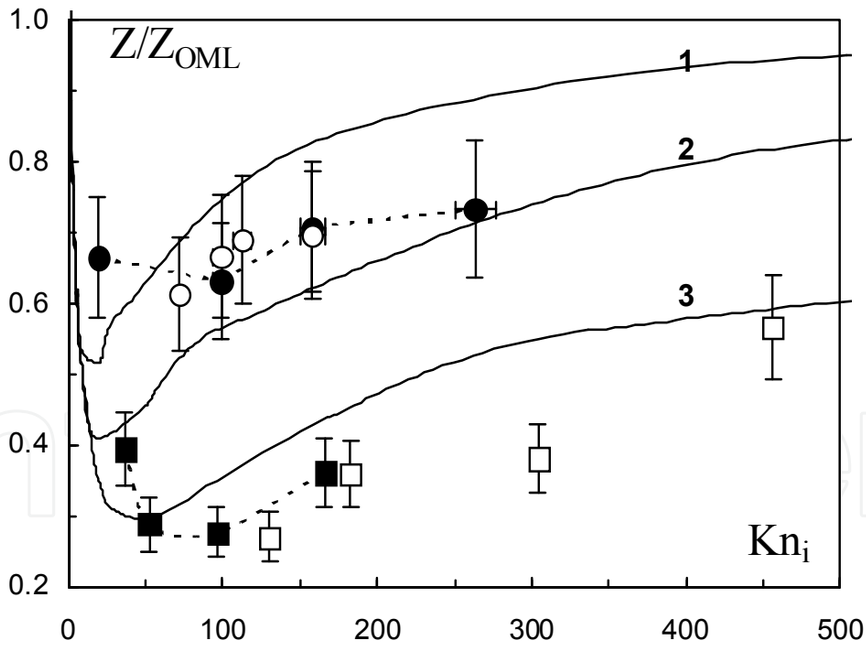


Fig. 14. Calculated dependence of Z/Z_{OML} (lines) on Kn_i for various a_p/λ: **1** – 0.07 ; **2** – 0.023 ; **3** – 0.007. Symbols are the Z_{min}/Z_{OML} value for the grains of radius a_p:: (○; ●)- 6.37 μm; (□; ■) - 2.755 μm; that form the dusty mono-layer (●; ■), or multi-layer structure (○; □).

The ratio of Z_{min} to the grain charge, $Z_{OML}\approx (2.7\ a_p T_e\ /e^2)$, obtained in the Orbital Motion Limited (OML) approach, are shown in Fig. 14 versus the Knudsen number $Kn_i = l_{in}/\ a_p$. Here T_e =3 eV is the electron temperature typical for rf-discharge in argon (Raizer, 1991;

Raizer et al., 1995), $l_{in} = (8\ T_i/\{\pi\ m_i\ \nu_{in}^2\})^{1/2}$ is an ion mean free path between their collisions with the gas neutrals, ν_{in} [s^{-1}] $\cong 8\ 10^6\ P$ [Torr] is the effective frequency of these collisions for single-charged argon ions (Raizer, 1991; Raizer et al., 1995), $T_i \cong 0.026$ eV is their temperature and m_i is the mass. The results of numerical simulations (Vaulina et al., 2006) of the charging of a single spherical particle in the weakly ionized argon-discharge plasma in the case of the neglecting of the drift ion velocity V_{id} are also presented in Fig. 14 as the ratio, Z/Z_{OML}, for the various values of a_p/λ = 0.07; 0.023; 0.007; here $\lambda \approx \lambda_{Di} = \{T_i/(4\pi e^2 n_i)\}^{1/2}$, and n_i is the concentration of ions (~10^7-5 10^8 cm^{-3}). A simple quantitative comparison of the presented numerical results, $Z(Kn_i)$, with the experimental data, $Z_{min}(Kn_i)$, is incorrect at least because of two reasons. First of them it is the neglecting of the dust screening, with a presence of which the obtained value of Z_{min} can be much lower than the real grain charge. The second reason is the neglecting of the drift ion velocity V_{id} in the numerical simulations (Vaulina et al., 2006) that may be not suitable for conditions of considered experiments in the near-electrode area of RF- discharge. In spite of that, we should note the good qualitative agreement between numerical $Z(Kn_i)$ and experimental dependencies $Z_{min}(Kn_i)$.

8. Conclusion

The results of numerical investigation of mass-transfer processes in extensive quasi-two-dimensional and three-dimensional non-ideal dissipative systems are presented. The particles in these systems were considered interacting with various isotropic pair potentials, which represented different combinations of power-law and exponential functions, commonly used for simulation of repulsion in kinetics of interacting particles. The calculations were performed in a wide range of parameters typical for the laboratory dusty plasma in gas discharges.

The evolution of mass-transfer processes, the velocity autocorrelation functions and the diffusion constants were studied. It was obtained that for the systems under study the particle temperature, the effective coupling parameter and the scaling parameter determine all mentioned characteristics. It was shown that the evolution of mean-square displacement of particles for the short observation times corresponds to the lattice oscillations with frequency proportional to the second derivative of pair potential of interparticle interaction. The estimations of the characteristic oscillation frequencies of particles (ω_t) in 3d- bcc-structures and in 2d- hexagonal lattice are presented. It is shown that these frequencies are responsible for the time of "settled life" of particles ($\tau_o \approx 2/\omega_t$) in non-ideal liquid systems and define the behavior of mass-transfer processes at the short observation times ($t < \tau_o$). The obtained results are in a good agreement with the "jumps" theory.

The results of the experimental study of mass-transfer processes are presented for the dust systems, forming in the laboratory plasma of radio-frequency (RF-) capacitive discharge. The experiments were carried out for macroparticles of various sizes ($a_p \approx 2.75$ μm and $a_p \approx 6.37$ μm) within a wide range of coupling parameters of dusty sub-system. The velocity autocorrelation functions, mass-transfer functions, the diffusion coefficients, pair correlation functions and the concentration were measured. For the most experiments, the motion of grains has the diffusive character. The diffusion coefficients, D, were determined from the Green-Kubo relation and from the mean-square displacement of particles. The difference between the values of D, obtained by two different methods, didn't exceed 5% and was within the limits of experimental error.

In all the cases there was obtained a good agreement between the direct measurements of the velocity autocorrelation and mass-transfer functions and those functions, calculated from the mean-square displacement of particles using the Eqs. (15a)-(15b). That means that the stochastic model, given by the system of Langevin equations, can be used for the correct description of the motion of dust under experimental conditions

The method of simultaneous determination of dusty plasma parameters, such as kinetic temperature of grains, their friction coefficient, and characteristic oscillation frequency is proposed. The parameters of dust were obtained by the best fitting the measured velocity autocorrelation and mass-transfer functions, and the corresponding analytical solutions for the harmonic oscillator. The coupling parameter of the systems under study and the minimal values of grain charges are estimated. The obtained parameters of the dusty sub-system (diffusion coefficients, pair correlation functions, charges and friction coefficients of the grains) are compared with the existing theoretical and numerical data.

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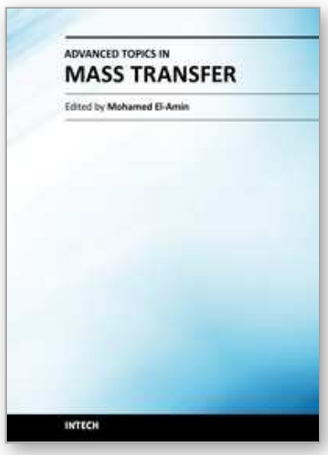

Advanced Topics in Mass Transfer

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