

# KINETIC THEORY OF NANOPARTICLES TRANSPORT AND RELAXATION PROCESSES IN GASES AND LIQUIDS

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**Abstract.** The present paper is devoted to nanoparticle relaxation and transport in fluids. These processes are considered in both rarefied gases and liquids. In the latter case, the molecular dynamics approach is used. On the other hand, a kinetic theory is constructed for nanoparticle transport processes in rarefied gases. In particular, the diffusion of nanoparticles in fluids and the effective viscosity of gas nanosuspensions are studied.

**Keywords:** Kinetic theory, nanoparticles, relaxation, transport processes, diffusion coefficient, viscosity of gas nanosuspensions, nanofluids.

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## 1. INTRODUCTION

Rapid development of nanotechnologies in the last two decades has defined the importance of studying nanoparticle transport processes in fluids. There are several different approaches to describing nanoparticle diffusion. Most investigators believe that the relaxation and transport of nanoparticles can be described in the same way as ordinary Brownian particles. On the other hand, attempts have been made to study the diffusion of nanoparticles using some kinetic theories, for example, a modified Enskog theory.

Strictly speaking, there are no grounds to describe the diffusion of nanoparticles in either of these ways. The nanoparticles size is of the order of the hydrodynamic infinitesimal scale. Therefore, we cannot use a hydrodynamic description. Nor can we apply Boltzmann's kinetic theory of molecular gases because the molecule–nanoparticle interaction is a collective one. Each gas molecule interacts simultaneously with all atoms (or molecules) of a nanoparticle. Nanoparticles are microscopic ones from a hydrodynamic point of view. However, these particles are larger than usual molecules. Therefore, gas nanosuspensions (gas + nanoparticles) and nanosuspensions (liquid + nanoparticles) differ significantly from gas mixtures, ordinary gas suspensions, and suspensions. The only way to study nanoparticle transport in fluids is to invoke nonequilibrium statistical mechanics and, in particular, kinetic theory.

The present paper gives a review of the studies of nanoparticle relaxation and transport in fluids performed recently by the author and his group. We consider these processes in both rarefied gases and liquids. In the latter case, the molecular dynamics method is used.

## 2. KINETIC EQUATIONS OF RAREFIED GAS NANO-SUSPENSIONS

A gas is defined as a rarefied one if the corresponding van der Waals parameter is small enough and if only binary collisions between its constituent molecules occur. Let a carrier gas and a pseudo-gas of nanoparticles be rarefied, i.e.,

$$\varepsilon_g = n_g r_0^3 \ll 1, \quad \varepsilon_p = n_p R^3 \ll 1. \quad (2.1)$$

Here  $r_0$ ,  $R$  and  $n_g$ ,  $n_p$  are the characteristic radii of the molecules and nanoparticles, respectively, and  $n_g$  and  $n_p$  are the densities of the carrier gas and the pseudo-gas of nanoparticles, respectively. In rarefied gases, the characteristic size of nanoparticles is much smaller than the mean free path of carrier gas molecules. Furthermore, the collisions of nanoparticles with carrier gas molecules are binary if the following condition is satisfied:

$$n_g R^3 = n_g (R/r_0)^3 \ll 1. \quad (2.2)$$

In this case, the dynamics of a gas nanosuspension is described by Boltzmann's equations [1, 2]

$$\frac{\partial f_g}{\partial t} + \mathbf{v}_g \cdot \frac{\partial f_g}{\partial \mathbf{r}} = J_B^{gg} + J_B^{gp}, \quad \frac{\partial f_p}{\partial t} + \mathbf{v}_p \cdot \frac{\partial f_p}{\partial \mathbf{r}} = J_B^{pp} + J_B^{pg}. \quad (2.3)$$

Here,  $f_g$  and  $f_p$  are the distribution functions of the carrier gas molecules and the pseudo-gas nanoparticles, respectively; the subscripts  $p$  and  $g$  refer to the particles and gas molecules, respectively. It is necessary to note that condition (2.2) is valid for all typical gas nanosuspensions if the carrier gas is not too dense.  $J_B^{gg}$ ,  $J_B^{pp}$ ,  $J_B^{gp}$ , and  $J_B^{pg}$  are the Boltzmann collision integrals for the molecules, particles, molecules and particles, and particles and molecules, respectively.

As noted in the Introduction, the interaction of a carrier gas molecule with a particle is collective and the molecule interacts simultaneously with all atoms (or molecules) of the particle. When this interaction is described by the Lennard-Jones 6–12 potential, the nanoparticle–molecule interaction is described by the following pair potential [3, 4]:

$$\Phi(r) = \Phi_9(r) - \Phi_3(r), \quad (2.4)$$

$$\Phi_i(r) = C_i \left[ \left( (r-R)^{-i} - (r+R)^{-i} \right) - a_i \left( (r-R)^{-(i-1)} - (r+R)^{-(i-1)} \right) \right].$$

Here,  $C_9 = 4\pi\epsilon_{ij}\sigma_{ij}^{12}/(45V)$ ,  $C_3 = 2\pi\epsilon_{ij}\sigma_{ij}^6/(3V)$ ,  $a_9 = 9/(8r)$ ,  $a_3 = 3/(2r)$ ,  $V$  is the effective volume per molecule of the dispersed particle,  $\epsilon_{ij}$  and  $\sigma_{ij}$  are the Lennard-Jones 6–12 potential parameters that describe the interaction between a molecule of the carrier gas and each molecule (atom) of the nanoparticle. Thus the molecule–particle interaction potential depends only on the parameters of the pair interaction potential for the carrier gas molecules and the dispersed-particle atoms ( $\epsilon_{ij}$ ,  $\sigma_{ij}$ ) and on the particle size.

### 3. DIFFUSION OF NANOPARTICLES IN RAREFIED GASES

As noted above, the Einstein–Langevin theory is commonly used to describe the diffusion of nanoparticles in dense gases and liquids. In this case, the diffusion coefficient is given by

$$D_E = kT / \gamma_s, \quad \gamma_s = 6\pi\eta R, \quad (3.1)$$

where  $T$  is the temperature of the medium and  $\eta$  is the viscosity of the carrier fluid. This formula is extended to rarefied carrier gases by using the Cunningham–Millikan–Davis (CMD) experimental correlation [5]

$$D_{KMD} = kT / \gamma_{KMD}, \quad (3.2)$$

here  $\gamma_{KMD} = 6\pi\eta R [1 + AKn_R + BKn_R \exp(-b/Kn_R)]^{-1} = \gamma_s / C$ ,  $A=1.257$ ,  $B=0.4$ ,  $b=1.1$ ,  $Kn_R = l/R$  is the Knudsen number. In practice, correlation (3.2) is included in all techniques for measuring the diffusivity of dispersed particles in rarefied gases [6].

On the other hand, attempts have been made to describe the diffusion of nanoparticles by means of the kinetic theory of gases. We mention only two of such attempts. The first was undertaken by Epstein [7]. He derived the following expression for the diffusion coefficient:

$$D_{Ep} = 3\sqrt{mkT} \left[ \rho R^2 \sqrt{2\pi} (8 + \alpha\pi) \right]^{-1}, \quad (3.3)$$

where  $m$  is the mass of the carrier gas molecule,  $\rho$  is the gas density and  $\alpha$  is the accommodation coefficient. There have been attempts to use the kinetic theory for hard spheres. The corresponding diffusion coefficient has the form [8]

$$D_{hs} = \frac{3\sqrt{2kTm(m+M)}}{16\rho(r_0 + R)^2\sqrt{\pi M}}, \quad (3.4)$$

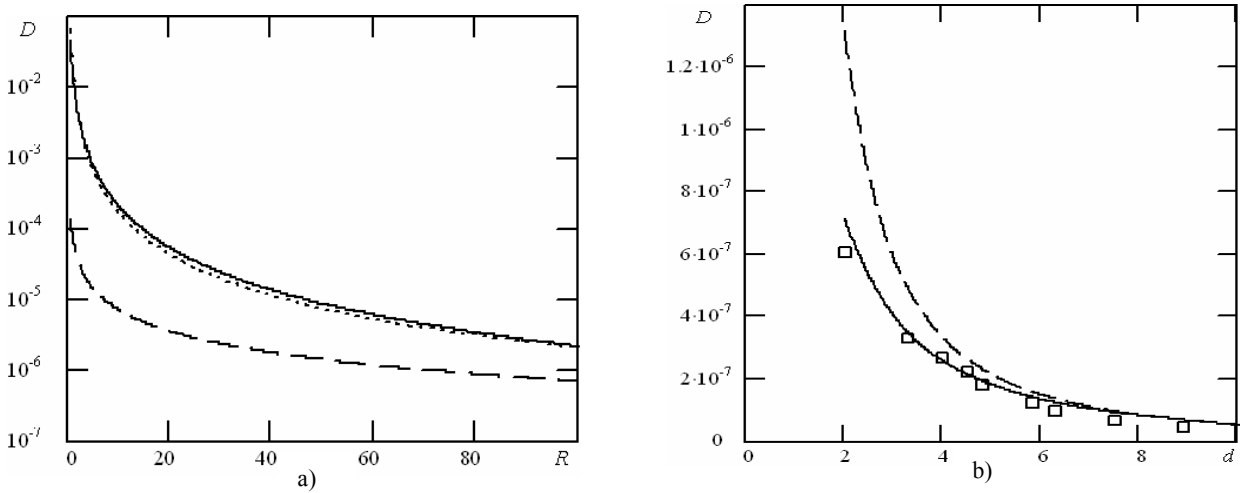
where  $M$  is the mass of the particle.

We have discussed the invalidity of using formulas (3.1) and (3.4). Expression (3.3) is semi-empirical. The accommodation coefficient included in it is not a universal constant and depends on the particle material, its temperature, etc. The Boltzmann kinetic equations (2.3) with the particle–molecule potential (2.4) give

$$D = \frac{3\sqrt{2kTm(m+M)}}{16\rho R^2\sqrt{\pi M}\Omega_{12}^{(1,1)*}(R)}. \quad (3.5)$$

Here  $\Omega_{kl}^{(i,j)*}$  is so-called  $\Omega$ -integral.

One of the most important characteristic features of the diffusivity of dispersed particles is its dependence on the particle radius. For fairly large particles,  $D \sim R^{-1}$  (see Eq. (3.1)). From Epstein's formula, we have  $D \sim R^{-2}$ . Our formula (3.5) gives a more complicated dependence [4, 9] because the  $\Omega$ -integral is also a function of the radius  $R$  too:  $D \sim R^{-2}(1 + a_1R^{-1/2} + a_2R^{-2})$ . Figure 1a shows a comparison of these dependences of the diffusion coefficients (3.5) ( $\text{cm}^2/\text{s}$ ) on the nanoparticle radius (nm) with the CMD correlation (3.2) and the Einstein formula (3.1) at a constant temperature  $T=300$  K.

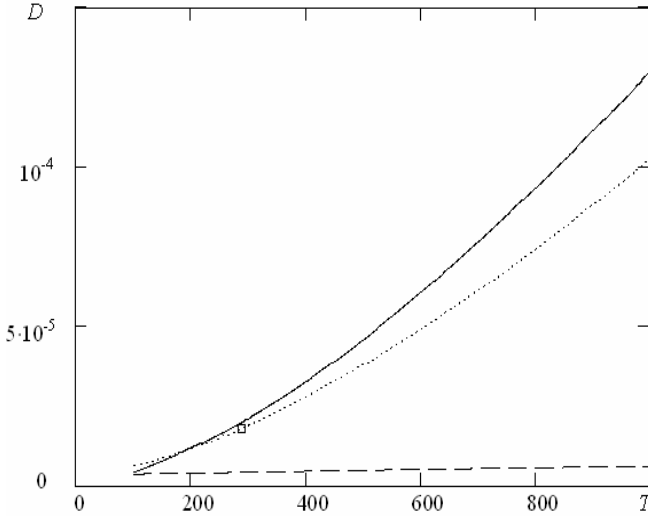


**FIGURE 1.** Diffusion coefficient of Zn in Ne nanoparticles vs. particle radius (Å); the solid curve refers to the kinetic theory (2.4), (3.5), the dotted curve to the CMD correlation, and the dashed curve to Einstein's formula. b) Diffusion coefficient of Zn in Ne vs. particle diameter (nm). The solid and dash curves are obtained from the kinetic theory (3.5), (2.4) and the CMD correlation (3.2), respectively. The squares are experimental data points.

As one might expect, the Einstein formula (3.1) is not applicable over the entire range of particle radii. At the same time, the CMD correlation coincides with our data for a nanoparticle size of up to 10 nm. However, for smaller particle sizes, the discrepancy between the kinetic theory and the CMD correlation is significant. For example, for particles with a characteristic size of about 2 nm, the CMD correlation gives a value for the diffusion coefficient which is more than twice that predicted by the kinetic theory.

To study the applicability of the kinetic theory and the CMD correlation to the diffusion of nanoparticles, we performed a special series of experiments. Particle diameters were measured by a differential mobility analyzer (DMA)

and an electron microscope (EM) [10] to obtain independent experimental data. In these experiments, we used nanoparticles of copper oxide ( $\text{Cu}_2\text{O}$ ) and nitrogen as the carrier gas. The experimental data obtained are presented in Fig. 1b (squares). It is evident that the results obtained from our kinetic theory are in good agreement with the



**FIGURE 2.** Diffusion coefficient  $D$  vs. temperature

experimental data. At the same time, the DMA method gives considerably overestimated values for the diffusion coefficient. This discrepancy becomes especially significant for particles of small sizes.

For Brownian particles, the temperature dependence of the diffusivity is very weak. For example, for particles in air,  $D_E \sim T^{0.35}$  [4]. At the same time, this dependence for nanoparticles, like that for molecules, is considerably stronger. The dependence of the diffusion coefficient of nanoparticles on temperature is especially strong for fine particles when the  $\Omega$ -integrals are different from unity. Figure 2 demonstrates the temperature dependence of the diffusion coefficient for condensation nuclei of fixed radius ( $R=30$  nm) in air at atmospheric pressure. The solid and dotted curves are obtained using the kinetic theory (3.5), (2.4) and the CMD correlation (3.2), respectively. The squares are experimental data [11]. The dashed curve corresponds to the Einstein formula (3.1).

Thus, the diffusivity of nanoparticles varies by a factor of ten as the temperature varies from 300 to 1000 K. The CMD correlation is applicable in a very narrow temperature range, which is not surprising. The constants of the CMD correlation (3.2) were determined in a narrow temperature interval (19-24° C). Thus, these constants are functions of the temperature of the carrier gas.

#### 4. EFFECTIVE VISCOSITY COEFFICIENT OF RAREFIED GAS NANOSUSPENSIONS

The presence of dispersed solid particles in a continuum radically changes its properties, including viscosity. The viscosity of suspensions with a low particle number density is usually calculated by the formula based on Einstein's phenomenological theory [12]. The viscosity coefficient calculated by this formula

$$\eta = \eta_0 [1 + (5/2)\varphi] \quad (4.1)$$

is higher than the viscosity of the carrier liquid  $\eta_0$ , here  $\varphi$  is the volume concentration of the dispersed particles. Later, this formula was extended to suspensions with higher particle concentrations (see, for example, [13, 14] and references therein). For rarefied gas suspensions, such approaches are absent. The aim of this section is to calculate the viscosity coefficient of rarefied gas suspensions using the kinetic theory with the molecule-particle potential (2.4). The particle-particle interaction will be described by the hard sphere potential [15].

We consider a rarefied gas nanosuspension as a binary mixture of carrier gas molecules and nanoparticles. It is easy to show that as a first approximation for the molar fraction of nanoparticles  $x_2$  the effective viscosity coefficient of gas nanosuspension takes the following form [15]

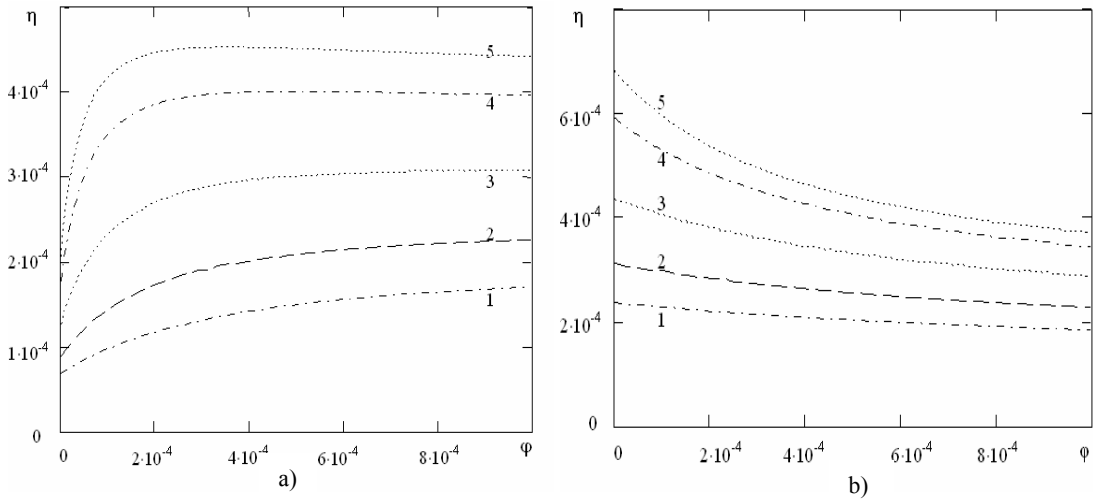
$$\eta = \eta_1 \left\{ 1 + \frac{x_2}{1 + 0.6A_{12}^* \mu} \left[ 0.3A_{12}^* \frac{(1 + \mu)^2}{\mu} \frac{\eta_{12}}{\eta_1} + 2 - 1.2A_{12}^* - 2 \frac{\eta_1}{\eta_{12}} \right] \right\}. \quad (4.2)$$

Here

$$\eta_i = \frac{5}{16} \frac{(\pi m_i kT)^{1/2}}{\pi \sigma_{ii}^2 \Omega_i^{(2,2)*}}, \quad \eta_{12} = \frac{5}{16} \frac{(\pi m_{12} kT)^{1/2}}{\pi \sigma_{12}^2 \Omega_{12}^{(2,2)*}},$$

subscripts 1 and 2 refer to the gas molecules and nanoparticles, respectively,  $m_1 = m$ ,  $m_2 = M$ ,  $m_{12} = mM/(m+M)$ ,  $\sigma_{22} = 2R$ ,  $\sigma_{11}$  and  $\sigma_{12}$  are the parameters of the Lennard-Jones potential for interactions between molecules and between particles and molecules.  $A_{12}^* = \Omega_{12}^{(2,2)*} / \Omega_{12}^{(1,1)*}$ .

The function (4.2) depends strongly on the mass ratio  $\mu = m/M \ll 1$ , nanoparticle radius, temperature, and the parameters of the potential (2.4). In particular, the function (4.2) can change sign for certain values of these quantities. This implies that the addition of small volume fractions of dispersed solid particles to the gas can both increase and decrease the effective viscosity of the medium. Thus, a decrease in the effective viscosity coefficient compared to the viscosity coefficient of the carrier gas  $\eta_1$  should be expected in the case of a large difference in size between the carrier gas molecules and nanoparticles. If the nanoparticle size is small enough, a decrease in the effective viscosity coefficient can be expected for gas nanosuspensions with a rather small density ratio of the particle material to the carrier gas.



**FIGURE 3.** Effective viscosity coefficient (in poise) vs. volume concentration of nanoparticles. a)  $U-H_2$  gas nanosuspension, and b)  $Zn-Ne$  gas nanosuspension.  $R = 0.5$  nm. Curve 1 corresponds to a temperature  $T=200$  K, curve 2 to 300 K, curve 3 to 500 K, curve 4 to 800 K, and curve 5 to 1000 K.

The example in Fig. 3 illustrates two different situations. The addition of a small amount of fine uranium particles to hydrogen increases the viscosity of the gas nanosuspension. In contrast, the effective viscosity of the  $H_2 - U$  gas nanosuspension with particles of sufficiently large radius  $R > 36$  nm is lower than that of the carrier gas [9]. On the other hand, the viscosity of the  $Zn-Ne$  gas nanosuspension is below that of pure neon. At room temperature and a particles volume concentration of  $2 \cdot 10^{-4}$ , the effective viscosity of this gas nanosuspension is approximately 15 % lower than the viscosity of pure neon, this effect being more pronounced with increasing temperature.

## 5. NANOPARTICLES RELAXATION IN DENSE GASES AND LIQUIDS

No kinetic equations are available to describe the dynamics of nanoparticles in dense gases and liquids where the characteristic particle size is comparable to or even larger than the free path length of the carrier gas molecules. In this case, the collision of a nanoparticle with gas molecules is a multiparticle process (the particle interacts simultaneously with several molecules of the medium). Moreover, one can show that for this case, it is in principle impossible to derive a kinetic equation for a one-particle distribution function. Such gas nanosuspensions are similar to non-ideal dense gases. In these cases, it seems natural to use molecular dynamics (MD) methods. This section gives a brief review of the results obtained with this technique. The hard-sphere model is applied to nanoparticles and carrier

fluid molecules because the medium is very dense and attractive interparticle forces are not as important as in rarefied media. The nanoparticle size is  $d = \sigma \div 5\sigma$ , and the nanoparticle mass is varied from  $M = m$  to  $M = 300m$  (here  $\sigma$  is the diameter of the carrier fluid molecule). The ordinary molecular dynamics algorithm is used for simulation. A cubic cell contains  $N$  molecules and one nanoparticle. The density of the medium is defined by the parameter  $\alpha = (V - V_p)/V_0$ , where  $V$  is the cell volume,  $V_p$  is the nanoparticle volume, and  $V_0 = N\sigma^3/\sqrt{2}$  is the close-packed volume for the molecules. The parameter  $\alpha$  is varied from 1.5 to 10.0. Periodic boundary conditions are used to compensate for the finite number of molecules in the cell.

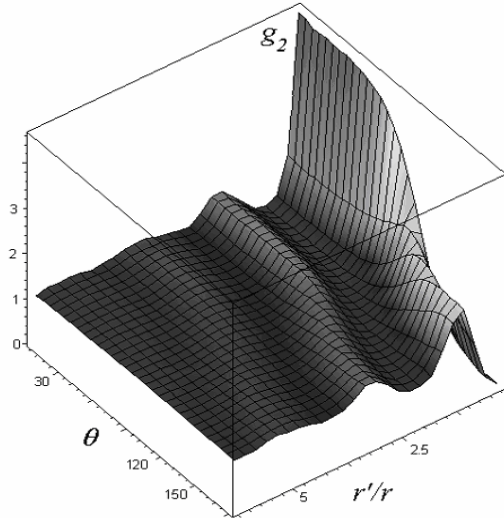
In rarefied carrier gases there is a single mechanism of nanoparticle relaxation. The relaxation is related to individual interactions of nanoparticles with molecules and has a characteristic time of the order of the mean free time of the molecules. The interaction of nanoparticles with a condensed carrier medium is of a much more complicated nature. The nanoparticle relaxation is characterized by the behaviour of the velocity autocorrelation function  $\chi_{vv}$  of the particle. MD calculations have shown that this function is well approximated by the sum of two exponents [17]

$$\chi_{vv} = b_1 \exp(-t/\tau_1) + b_2 \exp(-t/\tau_2). \quad (5.1)$$

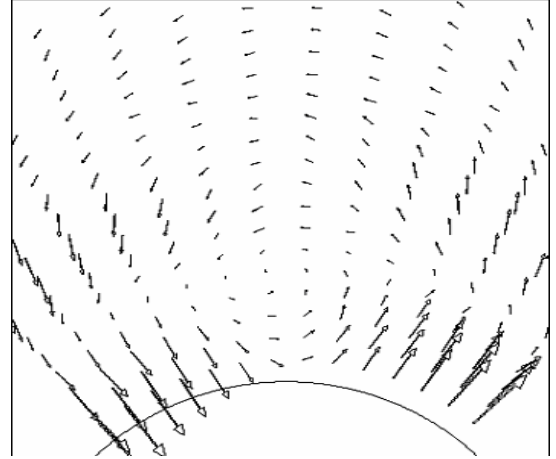
Here  $\tau_{1,2}$  are some relaxation times and  $b_{1,2}$  are coefficients. It has been shown that the processes of nanoparticle velocity relaxation are identical under equilibrium and nonequilibrium conditions [18, 19]. The first term in Eq. (5.1) is due to individual interactions of nanoparticles with molecules. However, the relaxation time  $\tau_1$  differs significantly from the Enskog relaxation time  $\tau_E$ . The ratio of the times  $\tau_1$  and  $\tau_E$  for various masses of nanoparticles is presented in Table 1 for a carrier medium density  $\alpha = 4.8$  and  $R/r = 4$ .

**TABLE 1.** The relation of the times  $\tau_1/\tau_E$ .

$M/m$	2	5	10	25	50	100	300
$\tau_1/\tau_E$	0.93	0.84	0.79	0.59	0.43	0.26	0.11



**FIGURE 4.** Radial distribution function of carrier fluid molecules near a nanoparticle



**FIGURE 5.** Flow field of the carrier fluid around a nanoparticle

The second term in Eq. 5.1 is due to the interaction of a nanoparticle with the nonequilibrium microfluctuations of the carrier-fluid density and momentum induced by the particle motion in the molecular medium. This interaction is the main mechanism of nanoparticle relaxation.

The microfluctuations induced by nanoparticles have a rather complex structure. They are characterized by local variations in the density and average velocity of the carrier fluid. The molecular density distribution near a particle is very anisotropic. In the example in Fig. 4 for the time  $t = 2\tau$  ( $\tau$  is the average mean free time of the molecules), the radial distribution function  $g_2$  of the molecules is presented as a function of the distance between the surfaces of particle and molecules and the angle  $\theta$  between the direction of particle motion and the line through centres of the particle and molecule. It can be seen that the density of molecules in front of the particle is several times higher than the average value. At the same time, behind the particle there is a region of low density. This disturbance propagates at the sound speed of the carrier fluid.

Local fluctuations in the average velocity of the carrier fluid have a great significance. The flow field around a nanoparticle has a toroidal vortex structure, which is shown in cross section in Fig. 5 (in the figure, the nanoparticle velocity is directed to the right). This structure has a characteristic size of the order of the nanoparticle diameter. One can see that the average velocity of the fluid near the particle has the same direction as the particle motion. Near the particle surface, this velocity has the maximum value. The resulting vortex accelerates the nanoparticle, thus retarding its relaxation.

The diffusion of nanoparticles in liquids and dense gases differ greatly from Brownian diffusion because there are two different mechanisms of nanoparticle relaxation. The diffusion coefficient of a nanoparticle is related to its velocity autocorrelation function by the Green–Kubo formula

$$D = \frac{1}{3} \int_0^{\infty} dt \chi(0, t).$$

Data obtained for various values of the mass ratio are given in Table 2, where  $\alpha = 4.8$  and  $R/r = 4$ . The values of the diffusion coefficient  $D_k$  obtained using the modified Enskog theory (which is actually a semi-empirical correlation) are given here for comparison. The Einstein diffusion coefficient  $D_E$  of Brownian particles is independent of the particle mass and  $D_E = 1.623$  for given density. The Enskog diffusion coefficient is almost always half the MD-calculated value.

**TABLE 2.** Diffusion coefficients of nanoparticles.

$M/m$	1	10	50	100	300
$D/D_E$	1.20	0.94	0.95	0.96	0.96
$D_k/D_E$	0.67	0.50	0.49	0.49	0.49

In contrast to the Einstein diffusion coefficient, which is almost independent of the carrier fluid density for  $\alpha > 20$ , the diffusion coefficient of nanoparticles increases linearly with the parameter  $\alpha$  as the density decreases.

## 7. CONCLUSIONS

Nanoparticles are diverse enough. They are properly nanoparticles, fullerenes, clusters, large organic molecules, etc. The transport properties of nanoparticles are very specific and are not adequately described by the models used for ordinary molecules or macroscopic dispersed particles (for example, Brownian particles). Essentially, we have a new physical object. To study its unique characteristics and properties, it is necessary to reject the traditional theoretical models and invoke the specific methods of obtaining information at the microscopic level.

A rigorous kinetic equation can only be derived for rarefied ultrafine and fine gas suspensions (see the classification of heterogeneous media in [2]). However, even in the simplest case of binary particle–molecule collisions, the interaction of carrier-gas molecules with a dispersed particle is collective and one needs to use the interaction potential (2.4). The corresponding kinetic theory describes the nanoparticle transport in rarefied gases very well. At the same time, the experimental CMD correlation for small particles is not absolutely adequate. This correlation is inapplicable for coarse particles at low and high temperatures. Therefore, in order to measure the diffusion coefficients and sizes of nanoparticles, it is necessary to develop new experimental procedures for treating experimental data.

The addition of nanoparticles to a rarefied gas can radically change its properties. In particular, the effective viscosity of gas nanosuspensions can be lower or higher than that of the pure gas. Physically, this behaviour is caused by a competition of several different mechanisms (the momentum transport by nanoparticles; a decrease in the molecule–molecule collision frequency due to collisions of molecules with nanoparticles; a reduction in the mole fraction of carrier-gas molecules, etc.).

The relaxation of nanoparticles in dense gases and liquids is radically different from the relaxation of both molecules and Brownian particles. In contrast to Brownian particles, in nanoparticle relaxation there is a kinetic stage. The main mechanism in this stage involves recollisions of the nanoparticle and molecules. However, the nanoparticle loses a relatively small fraction of its momentum in this stage. The main stage of relaxation is related to nanoparticle interactions with microfluctuations in the density and momentum of the carrier fluid. Similar processes are characteristic of Brownian particles but the relaxation time of nanoparticles is much smaller than that of Brownian particles. The second stage of nanoparticle relaxation is complex. It consists of a relaxation period of the drag force and a zone of its almost stationary value. The duration of the first period is of the order of  $20 \div 30$  mean free times of the carrier-fluid molecules. The second period is characterized by vortex structure formation in the carrier-fluid flow field. The subsequent evolution of nanoparticles is practically hydrodynamic. In this stage, the drag force acting on nanoparticles is close to the Stokes one if the nanoparticle mass is large enough [20] (see also the corresponding paper in this book).

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