

# Condensation and Temperature Forces between Aerosol Particles

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**Abstract.** The forces of interaction between aerosol particles induced as a result of condensation/evaporation processes or a particle-gas temperature (far away from the particles) difference are considered. The interaction is studied in the continuum, free-molecular, and transition regimes. The forces of interaction between two aerosol particles are determined using asymptotic and numerical methods (the DSMC method for solving the nonlinear kinetic Boltzmann equation and the statistical test particle method for solving the linear Boltzmann equation). The interaction forces are found as functions of the condensation regimes (values of Knudsen number), the distance between the particles and their shape, the vapor concentration, the condensation coefficient, and the particle surface temperatures. <sup>†</sup>

The possibility of particle coagulation is one of the significant properties of gas flows with solid or liquid particles. Many reasons leading to coagulation and processes accompanying the force interaction between the particles are well known. However, one of the reasons of particle collision, namely, vapor condensation on the particles, is not adequately studied. Mass- and heat-transfer processes during vapor condensation on the particles can lead to the force interaction between the particles (in particular, in the near aircraft wake). In the engine jets the particle dimension is small ( $10^{-9} \text{ m} \leq d \leq 10^{-5} \text{ m}$ , [1]); therefore, vapor can condense on the particles in the continuum, transition, and free-molecular regimes. Apparently, the condensation forces of interaction between two particles were first considered in [2]. In that study the forces acting on spherical particles located at an arbitrary distance were considered for the free-molecular condensation regime. In [3] the absolute value of the force of interaction between two spherical particles when the gas flow rates on their surfaces are specified (but not the evaporation, as asserted in the paper) was determined in the Stokes regime. The aim of the present study is to determine the condensation forces of interaction between aerosol particles over a wide range of the force parameters.

In the formulations of the problems studied we will use the following assumptions (justified, for example, for the majority of the particles in the aircraft trail): (1) all the processes are quasi-steady for the particles of the dimensions considered; the quasi-steady condensation regime is most rapidly established; and the particle approach time is less than the characteristic time of motion of the particles in the near trail; (2) the particle velocity with respect to the gas is negligible in considering the condensation and temperature forces; (3) the relative vapor concentration is significantly less than unity; (4) as a result of vapor condensation, the particle temperature increases only slightly; and (5) the electrostatic forces between the particles can be neglected.

## CONDENSATION FORCE

We will determine the force of interaction between two aerosol particles in a homogeneous gas mixture at a pressure  $p_\infty$  and a temperature  $T_\infty$ . One of the components of the gas mixture condenses on the particle surface. The condensation coefficient is equal to  $\alpha$  and the particle temperature is assumed to be uniform and equal to the gas temperature far away from the particles  $T_{wi} = T_\infty$ . The particle dimensions are comparable with the molecular mean free path. The adequate mathematic model for studying vapor condensation is the system of time-independent nonlinear kinetic Boltzmann equations for each component of the gas mixture. In the present paper we will use a

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simpler mathematic model, namely, two steady-state nonlinear kinetic equations for the condensable component and the background gas, respectively. The distribution functions of the condensable and background gases will be denoted by  $G$  and  $F$ . The system of kinetic equations for these functions is as follows:

$$\xi_i \frac{\partial F}{\partial x_i} = J(F, F) + J(F, G), \quad \xi_i \frac{\partial G}{\partial x_i} = J(G, G) + J(G, F) \quad (1)$$

Far away from the particles the boundary conditions are the following conditions of the equilibrium state of the gas:

$$G(x \rightarrow \infty) \rightarrow \varepsilon n_\infty M_G, \quad F(x \rightarrow \infty) \rightarrow (1 - \varepsilon) n_\infty M_F, \quad M(T, m_i) \equiv M_i = (h_i / \pi)^{3/2} \exp(-h_i \xi^2), \quad h_i = m_i / 2kT$$

Here  $\varepsilon = n_{G\infty} / n_\infty$  - is the relative number density (concentration) of the vapor and  $n_\infty = n_{F\infty} + n_{G\infty}$  - is the total number density of the gas. For the reflected molecules of the background gas we adopt the diffusion model of reflection from the particle surface. We will also assume that the vapor molecules are reflected from the particle surface with an equilibrium distribution function. The condensation and evaporation coefficients are assumed to be equal. The gas action force on a particle is determined by collisions of both vapor and background gas molecules with its surface  $F = F_G + F_F$ . If the vapor concentration is low ( $\varepsilon \ll 1$ ), the formulation of the problem is simplified. In the linear approximation in the small parameter the linear Boltzmann equation holds for the distribution function of the vapor molecules, while the linearized Boltzmann equation with a source term holds for the distribution function of the background gas molecules. In fact, the equilibrium distribution is the zero approximation of the distribution function of the background gas and the functions  $G_1$  and  $F_1$  must satisfy the following equations with the corresponding boundary conditions:

$$F = n_\infty M_\infty + \varepsilon F_1 + \dots, \quad G = \varepsilon G_1 + \dots, \quad \xi_i \frac{\partial G_1}{\partial x_i} = J(G_1, n_\infty M_\infty), \quad \xi_i \frac{\partial F_1}{\partial x_i} = L(F_1) + J(n_\infty M_\infty, G_1) \quad (2)$$

We note that for small Knudsen numbers the asymptotic solution of the kinetic equation (2) for the distribution function  $G_1$  reduces to the solution of the diffusion equation for the density  $n_G$  with the modified Hertz-Knudsen condition on the particle surface (see relations (4) below). For the function  $(G_1 + F_1)$  the asymptotic solution can be found as the solution of the Stokes equations with the boundary condition for the gas mass flow rate that is equal to the diffusion flow rate of the condensable vapor.

Within the framework of Eqs. (1), the problem was investigated numerically using the DSMC method. The masses of the vapor molecules and the background gas were assumed to be equal. All the molecular collision cross-sections were also assumed to be equal. The VHS interaction model with the potential  $\varphi \propto r^{-s}$  (where  $s=10$ ) was used. We investigated the forces of interaction between not only spherical particles but also particles of other shapes for a certain orientation of the particles, namely, hemispherical particles with the axis of symmetry perpendicular to the outward plane faces ( $L$  is the distance between the centers of the spherical parts of the surfaces); disk-shaped particles perpendicular to the axis of symmetry; cubic particles with parallel or perpendicular faces; and parallelepiped-particles with edges parallel to the axis line which are twice as great as the perpendicular edges of the parallelepipeds. In the last two cases the dimension  $L$  is the distance between the nearest faces.

At present there is no efficient method for solving the problem in formulation (2). Therefore, in the numerical investigation we used only the first of equations of system (2). In this case the solution obtained gives adequate results for large Knudsen numbers, while for the moderate Knudsen numbers we can calculate the most part of the interaction force. The first of equations of system (2) was solved using the well-known test particle Monte-Carlo method. In the free-molecular regime at moderate distances between the particles the particle interaction force was determined as the solution of integral equations by means the above-mentioned test particle method. In the continuum regime the problem of determination of the spherical-particle interaction force was also solved with allowance for the second of the equations of system (2). In this case the system consisting of two linear equations, namely, the diffusion equation for the vapor concentration and the Stokes equation for the velocity of the mixture, was solved using the method of separation of variables in the bispherical coordinate system.

## Analytical Results in the Free-Molecular and Continuum Regimes at Large Distances.

Let us study the interaction force between two particles of arbitrary convex shapes in the free-molecular regime. Their surface temperatures are uniform and equal to the gas temperature far away from the particles. Let  $p_{G\infty}$  - be the vapor pressure at large distances and  $p_{Gw}$  - be the saturated vapor pressure at the surface temperature. The surface condensation coefficients will be assumed to be different but uniform over the surface (in order to have the

possibility to simulate the interaction between overcooled drops and ice crystals). In the asymptotic approximation (at large distances  $L$ , subscript "as") the force is equal to

$$F_{as} = \frac{P_{Gw} - P_{G\infty}}{8\pi L^2} \left\{ \alpha_1 S_{1sh} [3S_{2sh} + 2(1-\alpha_2)S_{2eff}] + \alpha_2 S_{2sh} [3S_{1sh} + 2(1-\alpha_1)S_{1eff}] \right\} \quad (3)$$

$$S_{ish} = \iint_{(n_i e_x) > 0} (n_i e_x) dS_i, \quad S_{ieff} = \iint_{(n_i e_x) < 0} (n_i e_x)^2 dS_i$$

Here,  $n$  is the outward normal and  $x$  is the axis directed from particle 1 to particle 2.

We will now represent the asymptotic expression for the interaction force in the continuum limit at large distances between the particles only for spherical particles. In order to compare the continuum data with the results of the numerical solution we will use not only the expressions for the total interaction force but also the component  $G$ -of the force determined by vapor molecule-particle surface collisions. In this case we will consider both the values of the condensation coefficient of order of unity and small values of order of the Knudsen number. Usually (when the condensation coefficient is assumed to be of the order of unity), in investigating condensation on large particles the boundary condition of equality of the vapor concentration in the neighborhood of a particle and the saturated vapor concentration at the particle surface temperature is used. In this approximation the interaction force vanishes. Therefore, now on the outer boundary of the thin Knudsen layer adjacent to the surface of each particle we impose the generalized Hertz-Knudsen boundary condition for the vapor molecules:

$$j_G = \frac{c}{4} (n_{Gw} - n_G(r = R_i)) \frac{\alpha_i}{1 - b\alpha_i} = -D \frac{\partial n_G}{\partial r}, \quad n_G(r_i \rightarrow \infty) \rightarrow n_{G\infty} \quad (4)$$

Here,  $D$  – the vapor molecule diffusion coefficient in the background gas and  $c$  – the mean molecular velocity in equilibrium gas. In the generalized Hertz-Knudsen boundary condition the value of the coefficient  $b=0.432$  was borrowed from [4].

For  $R_1=R_2$ ,  $\alpha_1=\alpha_2$ , and at large distances between the particles the condensation force of attraction of spherical particles caused by collisions with only the vapor molecules was determined using the matched asymptotic expansions method. Its value is equal to

$$|F_G| = F_{G0} \frac{R^2}{L^2} \frac{4d}{(1+d)(1+2d)}, \quad F_{G0} = (p_{G\infty} - p_{Gw})\pi R^2, \quad d = \frac{4(1-b\alpha)}{\alpha c R} D = \frac{3\pi}{4} \frac{1-b\alpha}{\alpha} Kn \quad (5)$$

In the investigation of the above-mentioned component of the interaction force the similarity parameter is not the Knudsen number but a complex combination (variable  $d$ ) of the Knudsen number and the condensation coefficient.

The dependence of the force  $F_G$  on the parameter  $d$  is nonmonotonic and maximum is reached at  $d = d_m = \sqrt{2}/2$ .

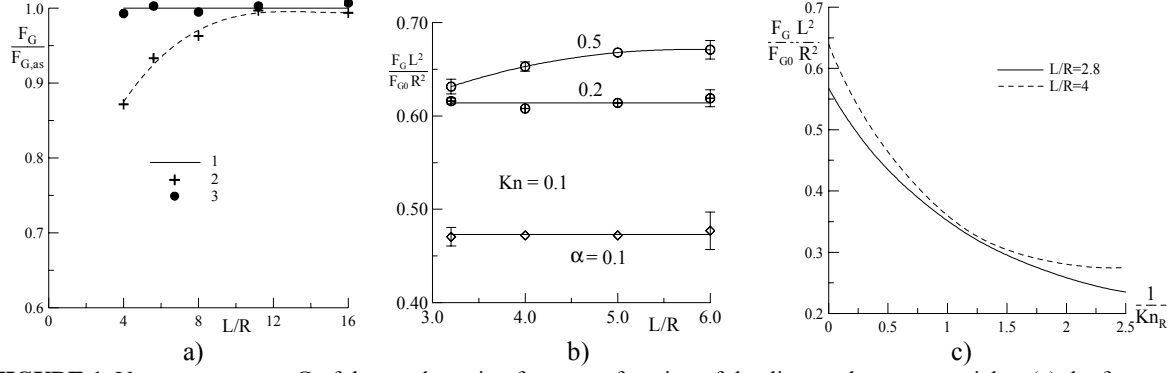
The total force of mutual attraction of two spherical particles of the same dimension determined by the vapor and background gas molecules collisions with particle is equal to

$$F^{cond} = F_{G0} \frac{45\pi}{32} \left( \frac{R}{L} Kn \right)^2 \frac{2d}{(1+d)(1+2d)} \quad (6)$$

We note that the total condensation force (6) decreases monotonically with the Knudsen number, this force depending nonmonotonically on the condensation coefficient.

## Comparative Analysis of the Results of Investigating the Condensation Force

In the free-molecular and continuum regimes the principal terms of the asymptotic expansion of the condensation interaction force (at large distances  $L$  between the particles) are inversely proportional to the second power of the distance. In free-molecular regime the error in this approximation was investigated in [2] for spherical particles. In Fig. 1 we have shown the error in this dependence for particles of various shapes in the free-molecular regime and for spherical particles in the transition regime (for the vapor component  $G$  of the interaction force). These data indicate that the condensation force is approximately inversely proportional to the second power of the distance between the particles in any condensation regime if the distances between the particles are not too small. Therefore, the distance  $L$  between the particles was taken to be equal to  $4R$  in order to analyze the dependence of the vapor component of the condensation force on the flow regime and the condensation coefficient. For spherical particles their dimensions and the condensation coefficients on their surfaces are assumed to be the same. In Fig. 2a we have

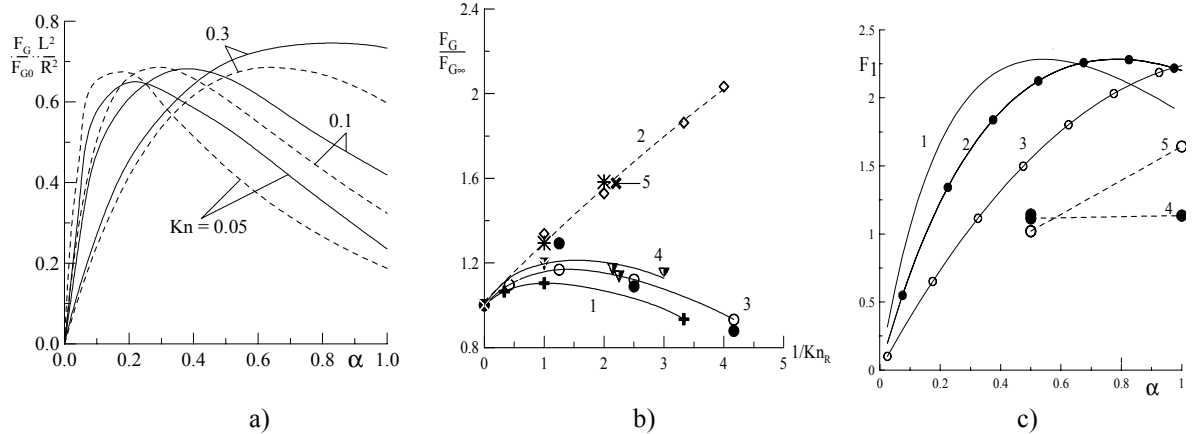


**FIGURE 1.** Vapor component  $G$  of the condensation force as a function of the distance between particles: (a) the free-molecular regime (1 corresponds to spheres, 2 to hemispheres, and 3 to disks); (b) the force acting on spherical particles in the transition regime for various condensation coefficients; and (c) the force acting on disk-shaped particles as a function of the inverse Knudsen number: curves 1 and 2 correspond to  $L/R=2.8$  and  $L/R=4$ , respectively

reproduced the results of the numerical simulation of the linear Boltzmann equation (see Eq. (2) for the distribution function  $G$ ) and compared these data with the asymptotic dependences (5) in the continuum condensation regime.

From these results (Fig. 2a) it follows that the nonmonotonic dependences of the vapor component of the condensation force on the condensation coefficient and the Knudsen number may be adequately approximated by means of the continuum dependences (for Knudsen numbers less than 0.3). These results were obtained in the linear approximation with respect to the vapor concentration. They strongly differ from the dependence for the pure vapor. In order to determine the error in the linear approximation for the vapor component of the condensation force we used the results obtained by means of the DSMC method in solving the nonlinear Boltzmann equation. In Fig. 2b we have plotted the graphs of the vapor component of the condensation force of interaction between spherical particles of the same dimension as a function of the inverse Knudsen number and the vapor concentration far away from the particles in the case of the condensation coefficient  $\alpha=1$ .

Our attempts to determine the value of the condensation force using the DSMC method for the smaller condensation coefficient failed even for moderate Knudsen numbers due to increases in the calculation error. The possibility to determine the total condensation force using the DSMC method may be estimated from the data given in Fig. 2c. From the data presented in Fig. 2c we can additionally conclude that the value of the condensation force estimated by means of the continuum approximation for Knudsen numbers greater than 0.3 and a small condensation coefficient can strongly differ from the real value.



**FIGURE 2.** Vapor component  $G$  of the condensation force of interaction between spherical particles in the transition regime: (a) as a function of the condensation coefficient and the Knudsen number;  $L=4R$ ; the results of the numerical and asymptotic continuum solutions of Eqs. (2) and (5) (solid and dashed curves, respectively); (b) as a function of the inverse Knudsen number for various vapor concentrations, 1 corresponds to the approximation linear with respect to the vapor concentration, 2 to the pure-vapor approximation, and 3, 4, and 5 to finite ratios of the vapor and background gas concentrations:  $n_G/n_F=0.25, 0.5$ , and  $10.0$ , respectively;  $\alpha=1$ ; and  $F_{G0}$  is the free-molecular value of the condensation force; (c) reduced condensation force  $F_1=F/[F_{G0} Kn^2 (R/L)^2]$  as a function of the condensation coefficient;  $L=4R$ ; 1, 2, 3 correspond to the continuum data; 4, 5 to the kinetic (DSMC) data; the Knudsen numbers: 1 correspond to  $Kn=0.24$ ; 2 and 4 to  $Kn=0.4$ ; and 3 and 5 to  $Kn=0.8$ .

In Fig. 3a we have plotted the graphs of the condensation interaction force as a function of the Knudsen number for spherical particles and in Fig. 3b for particles of various shapes. The interaction force is divided by its free-molecular value. We note that the error in calculating the background component of the condensation force is much greater than the error in calculating the vapor component (statistical error in both components was determined in the process of calculations). For Knudsen numbers less than 0.3 the asymptotic continuum approximation (6) for the total force of interaction between spherical particles leads to a reasonable error.

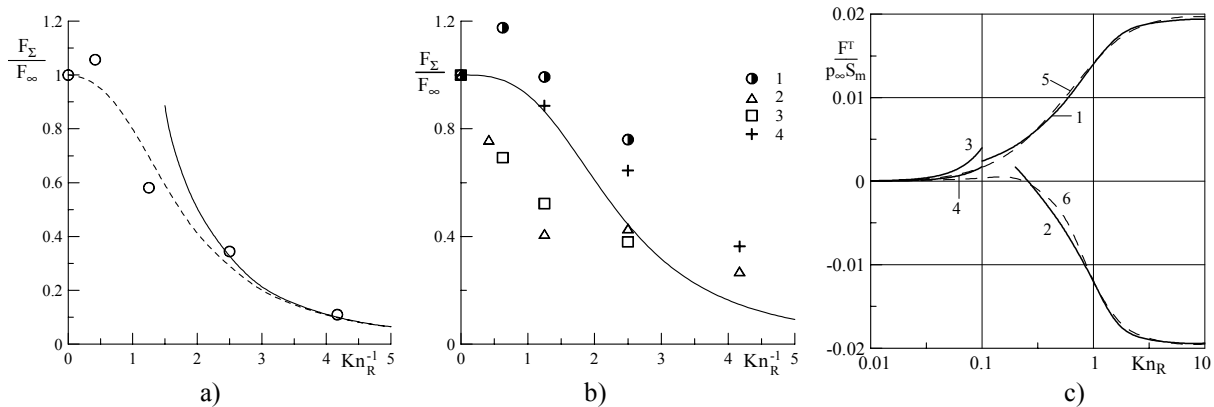
In what follows, we will propose an approximate expression for the condensation force. It is based on semi-empirical considerations and the asymptotic dependence (6) obtained in the continuum regime at large distances between the particles. At finite distances the error of expression (6) was determined by solving the Stokes equations with the generalized Hertz-Knudsen boundary conditions. When  $L/R=4$  the error in the asymptotic expression can reach 50%.

In order to calculate the effect of the condensation interaction force on the collision cross-sections of particles of an arbitrary dimension it is necessary to know the dependences of the condensation force for given values of the Knudsen number. For the spherical particles we can use the generalized Sherman approximation in which the continuum ( $F_{cond} \equiv F_c$ ) and free-molecular ( $F_{as} \equiv F_\infty$ ) approximations (6) and (3) are employed (see Fig. 3a)

$$[F_{app,1}(Kn)/F_\infty]^{-1.5} = 1 + [F_c(Kn)/F_\infty]^{-1.5} \quad (7)$$

In Fig. 3b the data scattering is due to the differences between the dependences of the total interaction force on the Knudsen number for particles of various shapes. Since the shapes of ice crystals may be significantly different, in this case it seems natural to use the approximation of the total interaction force based on the data concerning the particles of various shapes for determining the particle collision cross-section (see Fig. 3b):

$$F_{app,2}(Kn) = F_\infty f(Kn) \quad , \quad f = (1 + 0.08 \cdot Kn_R^{-3})^{-1} \quad (8)$$



**FIGURE 3.** (a,b) Total condensation force as a function of the inverse Knudsen number for  $\alpha=1$ : (a) for two spherical particles of the same dimension;  $L=4R$ ; the solid curve corresponds to the asymptotic dependence (6) and the dashed curve to the generalized Sherman approximation (7); (b) for particles of various shapes: 1 corresponds to hemispheres, 2 to disks, 3 to cubes, and 4 to parallelepipeds; and the solid curve corresponds to approximation (8). (c) Temperature force between spherical particles as a function of the Knudsen number (the value of the force is divided by the product of the gas pressure and the cross-sectional area of a particle): curves 1 and 2 correspond to the numerical results for the hot and cold particles, respectively; curve 3 and 4 to the continuum data in approximation (11); and curves 5 and 6 to approximations for any regime.

## TEMPERATURE INTERACTION FORCE

The processes of particle condensation and evaporation can lead to the onset of a difference between the surface temperatures of aerosol particles or the temperatures of drops and ambient gas. This temperature difference may also lead to the onset of the interaction force between small particles. In order to clarify the main properties of the temperature interaction forces, in particular, their dependence on the particle dimension (or on the gas rarefaction, i.e. the Knudsen number) we will study the temperature forces regardless of the condensation forces.

In [2] the temperature force of interaction between spherical particles located at a large distance  $L$  was determined for the free-molecular regime when the particle surface temperatures are uniform. Under these assumptions the  $j$ th particle acts on the  $i$ th particle with the force

$$F_{i,j}^{T, fm} (Kn \rightarrow \infty) = p_{\infty} \frac{3}{4} \pi \frac{R_i^2 R_j^2}{L^2} \left( \sqrt{\frac{T_{wj}}{T_{\infty}}} - 1 \right) \equiv \frac{3}{4} (p_{\infty} \pi R_i^2) \frac{R_j^2}{L^2} \left( \sqrt{\frac{T_{wj}}{T_{\infty}}} - 1 \right) \quad (10)$$

If the temperature of a particle is lower than the gas temperature then an attracting force acts on the other particle.

In the continuum regime and when the surface temperature is uniform the interaction force is due to the action of temperature stresses in the gas. Usually, it is assumed that only the viscous shear stresses can exist in a perfect gas in addition to the pressure. However, using kinetic gas theory, initially Maxwell and then Burnett have demonstrated that the temperature stresses can exist in the gas even in the continuum flow regime. In the developed asymptotic theory of slow nonisothermal gas flows [5] it was shown that an interaction force between two spherical nonisothermal particles exists when their surface temperature is uniform. When the difference between the particle and gas temperatures is small the value of the temperature force exerted on the  $i$ th spherical particle by the  $j$ th spherical particle is given by [6]:

$$F_{i,j}^T (Kn \rightarrow 0) = k_T \left( \frac{25}{16} \pi \right) p_{\infty} (\pi R_i^2) \frac{R_i R_j}{L^2} \left( \frac{T_{wi}}{T_{\infty}} - 1 \right) \left( \frac{T_{wj}}{T_{\infty}} - 1 \right) Kn_R^2 \quad (11)$$

In the continuum regime the direction of action of the forces differs from the direction in the free-molecular regime: the particles repulse one another when their temperatures are simultaneously higher or lower than the ambient gas temperature.

In order to determine the effect of the finite distance between the particles on the continuum value of the temperature interaction force we solved the equations of slow nonisothermal gas flows for  $L=4R$ . The value obtained differs by not more than 30% from the asymptotic expression (11).

In Fig. 3c we have reproduced the variation of the temperature force determined by the DSMC method in the transition regime. The force acting between two heated or cooled spherical particles of the same dimension is shown. The distance between the centers of the particles was equal to  $L=4R$ . In order to reduce the calculation error the surface temperature was taken to be twice as great as the gas temperature (for heated particles) and equal to  $(2-\sqrt{2})^2 T_{\infty} \approx 0.3432 T_{\infty}$  (for cooled particles). The choice of the latter value is attributable to the fact that in the free-molecular regime ( $Kn \rightarrow \infty$ ) at such a temperature the forces of interaction between the heated and cooled particles have the same magnitude (but are oppositely directed).

In Fig. 3c the negative values of the force correspond to the attraction forces. In this figure we have also shown the continuum approximations (11) for Knudsen numbers less than 0.1. As can be seen from the results found using the DSMC method for solving the Boltzmann equations in the transition regime, the prediction of asymptotic theory of slow nonisothermal gas flows is valid for small Knudsen numbers. This means that the interaction force rapidly decreases with the Knudsen number (proportionally to  $Kn^2$  at small  $Kn$ ).

**Conclusions.** Vapor condensation on aerosol particles leads to the little-known and weakly investigated effect of the onset of forces of interaction between neighboring aerosol particles. These interaction forces lead to increase in the particle collision cross-section as compared with the geometric cross-section and to increase in particle coagulation (in particular, in the near aircraft wake). The interaction force consists of two components: the condensation force which depends only on the mass flow to the particles, and the temperature force determined by the difference between the particle surface and gas temperatures. For large particles there are no such forces (when condensation occurs in the continuum regime). It is shown that the continuum equations (Navier-Stokes equations) with the generalized Hertz-Knudsen boundary conditions make it possible to determine the value of the condensation interaction force for Knudsen numbers less than  $Kn=0.3$ . The analysis of the force interaction performed should be generalized to include the case of the conjugate mass- and heat-transfer.

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