About a hypothesis of aerosol gravito-photophoresis in the atmosphere and its experimental verification

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The results of theoretical and experimental investigations of so-called accommodation forces (gravito-photophoresis) acting on particles with an asymmetry of surface properties in a rarefied gas in the directed electromagnetic radiation field are presented. The gas-kinetic theory of the phenomenon in the free-molecular regime predicts an occurrence of an unusual force, caused by the difference in accommodation coefficients of the normal pulse (momentum) on model particle hemispheres, which can be comparable with radiometric photophoretic forces for micron-size particles in stratosphere. The measured values of the force for steel particle in helium by a technique of model thermal-physics experiment agree well with theoretical predictions. The ratio of accommodation force to the radiometric photophoretic force in experiment does not exceed 3%.

Introduction

Noticeable manifestations of individual transport characteristics of particles, which are not reduced only to the convective aerosol transfer due to the tropospheric circulation, should be observed in thermally and mechanically stable stratosphere.[1,2] In the radiation field, vertical forces of different physical nature and magnitude can affect the particles of stratospheric aerosol of diverse origin, chemical composition, and morphology. It is well known that the ponderomotive forces (forces of light pressure) are decisive for the aerosol dynamics in intense laser beams, but are negligible for aerosol particles in the field of solar radiation.[3] At low gas pressures and low intensities of incident radiation, the vertical thermoconvective forces, produced by carrying away heated particles by surrounding gas volumes, are not significant.[4]

The forces of radiometric photophoresis are traditional in the analysis of the stratospheric aerosol levitation. In particular, in recent publications[5–7] the results are presented on calculations of characteristics of the photophoretic motion of particles (primarily, soot particles) at altitudes of lower and middle stratosphere. Together with results from Ref. 8, the obtained data enable one to assess in a new way the potentialities of the radiometric photophoresis in the absorbing aerosol vertical transport in the middle atmosphere. The quantitative measurements of the soot particle photophoresis rate[9] have posed the problem of developing the photophoresis theory for the fractal-like aggregates, which takes into account their unusual optical and gas-kinetic characteristics. Nevertheless, the question about one more possible class of extraordinary forces, affecting aerosols in the radiation field, remains controversial.

In 1951, F. Ehrenhaft and E. Reeger (Austria) have reported about observations of a so-called "transverse" photophoresis of particles of a fine graphite powder in argon at low pressures (7–13 hPa). Being illuminated by a horizontal beam of solar radiation, the particles demonstrated various motion trajectories including a vertical rise opposite to the gravity.[10] The discovered phenomenon (appearance of the force and the positive vertical velocity of the particle motion in the field of arbitrarily directed radiation) was called gravito-photophoresis.

Results of qualitative experimental investigations of this phenomenon were described in Refs. 11–13 for the motion of particles of different matters (powders of minerals, metals, graphite, carbon, volcanic aerosol, and so on) at their horizontal illumination by a halogen lamp with the radiation intensity close to the solar constant; the air pressure in the measurement chamber corresponded to tropospheric, stratospheric, and mesospheric altitudes. A portion of particles, demonstrating in the experiments the transverse gravito-photophoresis, was 0.01–1% of the whole number; the velocity of vertical motion of particles opposite to the gravity was several mm/s at sufficiently low air pressures. Later on, the same measurements were conducted at a modernized experimental setup, which enabled the control of the particle size, radiation intensity and direction, as well as studying the motion trajectories with a video camera.[14–15]

Carbon particles of up to 2 μm in diameter were used in the experiments; a xenon lamp provided for a wide range of radiation intensities (from one to three solar constants) in the spectral range close to the solar one. The air pressure in the measurement chamber varied in ranges corresponding to the stratospheric altitudes.

About 5% of carbon particles demonstrated the transverse gravito-photophoresis, the remaining particles demonstrated the longitudinal positive radiometric photophoresis.[15] The processing of the

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motion trajectories has made it possible to assess quantitatively the velocities of the particle vertical motion. It was demonstrated[15] that at the radiation intensity equal to the solar constant, the rate of the gravito-photophoresis for carbon particles was 0.6 mm/s (that is comparable with the rate of their gravitational sedimentation) and decreased at the pressure lowering.

Theoretical description of the observed results is based on the semiempirical theory of gravito-photophoresis, which has been developed by H. Rohatschek over several years. In this theory the phenomenological treatment of the appeared vertical force at an arbitrary direction of the incident radiation is described in terms of so-called body-fixed forces, in contrast to the space-fixed forces.[12,16,17] A simplified model of the physical mechanism initiating the gravito-photophoretic force is as follows.[11,12,17–19]

A spherical particle of high heat conductivity is in the directed radiation field. Its temperature differs from the temperature of the ambient gas at the cost of absorption, however, is homogeneous (in this case the radiometric photophoresis force, which is formed by the nonhomogeneity of the surface temperature, is negligible[20]). Particle hemispheres, frontal to radiation and the rear one, are characterized by asymmetry of surface properties, expressed in the difference of accommodation coefficients of molecular characteristics on the particle surface.

To calculate the appearing accommodation force, results of solution of the problem on the force, acting in a rarefied gas to a heated thin plate, characterized by different accommodation properties of its surfaces (the problem of a plate radiometer), are used. This problem was analyzed by M. Knudsen in 1930 by the methods of elementary kinetic theory when developing the radiometric method of determination of energy accommodation coefficients.[21] The immediate application of that result to solving the spherical radiometer problem leads to a simple expression for the gravito-photophoretic force[11–12]

$$F_{\mathrm{gph}} = \frac{\pi}{4}\frac{R}{M}\rho_g R_{\mathrm{p}}^2 (T_S - T_{\mathrm{g}})\Delta\alpha_E, \qquad (1)$$

where $T_S - T_{\mathrm{g}}$ is the mean difference between temperatures of the particle and the ambient gas; $\Delta a_E = a_{E_2} - a_{E_1}$ is the difference of energy accommodation coefficients for particle's different hemispheres; R is the universal gas constant; M is the molar mass; p_g is the air density; R_{p} is the particle radius. By the method of its derivation, expression (1) is true for conditions of the free-molecular regime, which almost without limitations is realized for submicron and micron particles in stratosphere and mesosphere. At a later time, the author of the theory of gravito-photophoresis tried to generalize that result to other gas-kinetic regimes (viscous-glancing and intermediate).[17–19]

The obtained theoretical results predict a high efficiency of gravito-photophoretic forces in vertical transfer of stratospheric and mesospheric particles against the gravity, with which no one of the known power mechanisms, inherent to atmospheric aerosols in the field of directed radiation (including radiometric photophoresis) can compete. This theory, for example, was proposed for explanation of the observed phenomenon of accumulation of soot particles from the aircraft engines at altitudes of the middle stratosphere.[19,22]

At present, this theoretical model without some basic corrections is used by other researchers in calculation of forces for more complex particles. They again came to conclusion about very high transport efficiency of the above power mechanism in the stratosphere and mesosphere and used it for explanation of many peculiarities of the aerosol spatial distribution in the middle atmosphere.[23–25] These results made them to analyze critically the initial positions, methods of analysis, and conclusions of the theory. Besides, there appeared a necessity of independent experimental study of the gravito-photophoretic forces by methods, different from the well-known ones.[11–15]

The goal of this paper is, first, the independent development of gas-kinetic theory of the phenomenon in the free-molecular regime and, second, an attempt of measuring the predicted forces using methods of model thermal experiment with macro-particles for quantitative comparison of results with theoretical predictions.

1. Gas kinetic theory of accommodation forces in the radiation field

Consider a spherical particle of the radius R_{p}, weighted in the unlimited volume of gas in the field of unidirectional electromagnetic radiation of the intensity I and the wavelength λ at the rate U_∞ of the unperturbed gas (Fig. 1), the particle thermal conductivity λ_{p} and the complex refractive index $m = n + ik$.

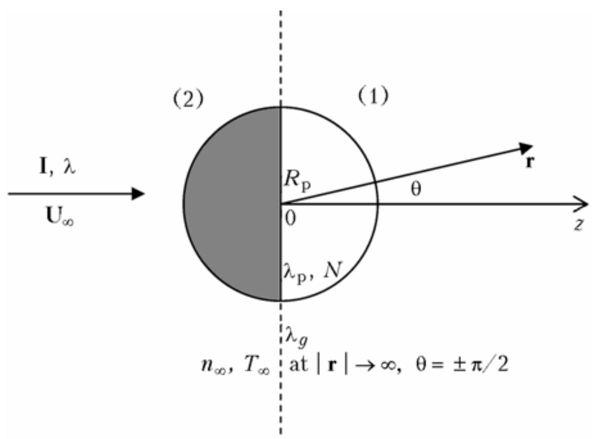

Fig. 1. Schematic view of the accommodation and photophoretic forces in the radiation field.

The particle has a model asymmetry of surface characteristics: frontal (further index (2), $\pi/2 \le \theta \le \pi$) and rear (index (1), $0 \le \theta < \pi/2$) hemispheres relative to the radiation direction, which are characterized by constant and yet various values of phenomenological accommodation coefficients of the pulse and energy of gas molecules. The volume thermo-physical and optical characteristics for both hemispheres are the same. Thus, the physical model of the particle fully corresponds to the model proposed in Refs. 17–19.

In conditions of free-molecular regime (the Knudsen number, $\mathrm{Kn} = l_g/R_p \gg 1$, where l_g is the mean free path-length of gas molecules) the distribution function for gas molecules, falling on the particle surface, can be taken as the Maxwellian distribution.

$$f^-(r = R_p) = f_\infty, \quad f_\infty = n_\infty\left(\frac{m}{2\pi kT_\infty}\right)^{3/2}\exp\left(-\frac{mv^2}{2kT_\infty}\right), \qquad (2)$$

where n_∞, T_∞ are the number density of molecules and the gas temperature far away from the particle at $|r| \to \infty$, $\theta = \pm\pi/2$; V is the velocity of molecules; m is the mass of a gas molecule; k is the Boltzmann constant. The distribution function f^+ of molecules reflected from the particle surface can be defined by different methods.[26] The method of physical modeling of the function is most widespread; it can be exemplified by the mirror-diffuse schematic diagram of Maxwell boundary conditions, where the share $(1 - \varepsilon)$ of falling molecules is scattered by the mirror surface, and the share ε is emitted in equilibrium at a local temperature of the surface element $T_s(\theta)$.

$$f^+(r = R_p) = (1-\varepsilon)f^-[\mathbf{v} - 2\mathbf{n}(\mathbf{nv})] + \varepsilon f^w, \qquad (3)$$

where

$$f^w(r = R_p) = f_\infty[1 + \nu_S(\theta) + (c^2 - 3/2)\tau_S(\theta)];$$

$$\nu_S = (n_S - n_\infty)/n_\infty, \quad |\nu_S| \ll 1;$$

$$\tau_S = (T_S - T_\infty)/T_\infty, \quad |\tau_S| \ll 1; \quad c = \left(\frac{m}{2kT_\infty}\right)^{1/2} v;$$

n_S and T_S are parameters of the linearized Maxwell function f^w [Ref. 26]; n is the outer normal to the particle surface. The known drawback of the scheme (3) is its single-parameter composition: the only model parameter is the coefficient of diffuse reflection ε, which determines the accommodation of any gas macro-parameter on the particle surface.

When solving a series of the fundamental problems of aerosol microphysics (the force of resistance,[27] thermo- and photophoresis of aerosol particles[28,20]), another simple model of boundary conditions, proposed independently by different authors,[29–31] can be used. In this model the distribution function of reflected molecules is modeled by the half-space series expansion of the local Maxwell distribution function

$$f^+(r = R_p) = f_\infty[1 + a_0(\theta) + a_{1n}(\theta)c_r +$$
$$+ a_{1\tau}(\theta)c_\theta + a_2(\theta)(c^2 - 3/2) + \ldots], \qquad (4)$$

where $a(\theta)$ denotes expansion coefficients depending on the polar angle θ for the radiation incidence on the particle surface. In the convergent series (4) the finite number of expansion terms remains due to the requirement of fulfilling basic conservation laws for transfer of macro-parameters.

The unknown coefficients of expansion are determined from relations of the balance of particle number fluxes, tangential and normal pulses, power of gas molecules at a local point on the particle surface. In writing the balance relations, the phenomenological accommodation coefficients in the so-called Knudsen (flux) determination[26] are used.

In none of the previously considered fundamental problems[20,27,28] the use of a given scheme of boundary conditions has resulted in physically contradictory results, and accommodation coefficients of pulse and energy, obtained from comparison with the experimental values are in good agreement with the known literature data for "technical," not atomic-clear, surfaces, that corresponds to conditions for real atmospheric aerosols.

In the final analysis, the coefficients of expansion $a_i(\theta)$ in Eq. (4) are functions of the accommodation coefficients of the tangentional pulse a_τ, the normal pulse a_n, and the energy a_ε of gas molecules on the particle surface, as well as the local temperature $T_S(\theta)$. Temperatures in the volume T_p and on the particle surface $T_S(\theta)$ are determined from the solution of a so-called "thermal" problem on the basis of the nonhomogeneous stationary equation of the thermal conductivity:

$$-\lambda_p \Delta T_p(r,\theta) = -\mathrm{div} I = 2n\kappa k_0 |I| B(x,\theta), \qquad (5)$$

where $B(x, \theta)$ is the function of sources of electromagnetic energy inside the particle[6]; $k_0 = 2\pi/\lambda$ is the wave number; $x = r/R_p$ is the dimensionless radial coordinate.

The solution for $T_S(\theta) = T_\infty(1 + \tau_S(\theta))$ has the form:

$$T_S(\theta) = T_\infty\left[1 + \frac{1}{4}\frac{IQ_{\text{abs}}}{f_2 p_\infty\sqrt{\dfrac{2kT_\infty}{\pi m}} + 4\sigma e T_\infty^4} + \right.$$
$$+ \frac{IJ_1}{f_2 p_\infty\sqrt{\dfrac{2kT_\infty}{\pi m}} + 4\sigma e T_\infty^4 + \dfrac{\lambda_p T_\infty}{R_p}}\cos\theta -$$
$$\left. - \sum_{l=2}^{\infty} P_l(\cos\theta)\frac{IJ_l}{f_2 p_\infty\sqrt{\dfrac{2kT_\infty}{\pi m}} + 4\sigma e T_\infty^4 + l\dfrac{\lambda_p T_\infty}{R_p}}\right], \qquad (6)$$

where

$$J_l = (2l+1)n\kappa\rho\int_0^{\pi}P_l(\cos\theta)\sin\theta\mathrm{d}\theta\int_0^1 x^{l+2}B(x,\theta)\mathrm{d}x;$$

$$\rho = 2\pi R_\mathrm{p}/\lambda;$$

$P_l(\cos\theta)$ are the Legendre polynomials; e is the emittance of the particle surface; σ is the Stefan–Boltzmann constant; f_2 is the accommodation complex (specific for different schemes of boundary conditions).

The second term in the right-hand side of Eq. (6) is stipulated by homogeneous heating of a particle at the cost of the radiation absorption (here Q_abs is the dimensionless factor of radiation absorption); the third term (proportional to the asymmetry factor of radiation absorption J_1 and $\cos\theta$) is connected with the radiometric photophoresis due to the temperature nonhomogeneity of the particle surface; the last term accounts for higher angular harmonics in the temperature distribution. Note that the useful approximation of a precise solution (6) in calculations is the cosinusoidal temperature distribution on the surface

$$T_S \approx T_\infty(1+\tau_{S0}+\tau_{S1}\cos\theta).$$

It is governed by a fast convergence of a series (6) because of the presence in the denominator of the fractions of the summand connected with the particle thermal conductivity. It should be also noted that the cosinusoidal distribution of the surface temperature of model macro-particles in the field of directed radiation was confirmed experimentally with a high accuracy.[32]

The force, affecting a stationary particle, is determined by integrating the total pulse flow of the gas molecule over its surface and is equal to the sum of the photophoretic and accommodation forces

$$F = -n_z\int_v \mathrm{d}v\sum_{i=1}^{2}\mathrm{d}S_i m\sum_{\mp}v_r v_z f^{\mp}(r=R_\mathrm{p},\theta,v) =$$

$$= F_\mathrm{ph}(\propto J_1) + F_\mathrm{ac}(\propto Q_\mathrm{abs}), \qquad (7)$$

where the indices $i = 1, 2$ correspond to rear and frontal hemispheres of a particle, and indices $\mp$ denote the falling and reflected flows of gas molecules, respectively. The photophoretic force for a model particle is of the form:

$$F_\mathrm{ph} = \frac{\pi}{6}R_\mathrm{p}^2 p_\infty I J_1\sum_i\frac{f_1(i)}{f_2(i)p_\infty\sqrt{\dfrac{2kT_\infty}{\pi m}}+4\sigma e(i)T_\infty^4+\dfrac{\lambda_\mathrm{p}T_\infty}{R_\mathrm{p}}}, \qquad (8)$$

where the accommodation complexes for the scheme of boundary conditions (3) are $f_1 = f_2 = \varepsilon$, and for the model (4)

$$f_1 = \frac{\alpha_E\alpha_n}{1-\dfrac{\pi}{32}(9-\alpha_E)(1-\alpha_n)},$$

$$f_2 = \alpha_E\frac{1-\dfrac{9\pi}{32}(1-\alpha_n)}{1-\dfrac{\pi}{32}(9-\alpha_E)(1-\alpha_n)}.$$

The limiting transition from Eq. (8) to a particle with surface-homogeneous characteristics gives the known result.[20]

The expression for the new accommodation force is of the form:

$$F_\mathrm{acc} = \frac{\pi}{16}R_\mathrm{p}^2 p_\infty I Q_\mathrm{abs}\sum_i(-1)^i\frac{f_1(i)}{f_2(i)p_\infty\sqrt{\dfrac{2kT_\infty}{\pi m}}+4\sigma e(i)T_\infty^4}, \qquad (9)$$

where the summation symbol, introduced for reduction of the equation, means the difference between two quantities with indices 1 and 2, corresponding to different accommodation characteristics of particle hemispheres. The term "accommodation force" here and below is used to denote a difference between new results and those obtained for the gravito-photophoretic force F_gph from Eq. (1); however, according to the author's model, the latter is also the accommodation force. At an identity of surface characteristics on hemispheres the accommodation force is equal to zero.

Note that the formation of this force is due to, first, the radiation heating of the particle and, second, the difference in the accommodation characteristics of particle hemispheres. The particle mean temperature is higher than the equilibrium gas temperature at the cost of the radiation absorption ($F_\mathrm{acc} \propto IQ_\mathrm{abs}$). Provided the radiation is absent, the temperature of the particle and gas is the same, and the presence of asymmetry of surface characteristics does not result in formation of the accommodation force. It follows from (9) that the accommodation force (as the photophoretic force) is proportional to the product of the accommodation coefficients of the energy and the normal pulse of gas molecules, more precisely, to the radiometric accommodation complex

$$\alpha_E\alpha_n/[1-\frac{\pi}{32}(9-\alpha_E)(1-\alpha_n)].$$

It is evident that in the absence of the accommodation of the power and the normal pulse, there are no photophoretic or accommodation forces on the particle surface. Note that the one-parametric mirror-diffuse scheme (3) does not clarify in detail the mechanism of origination of the above forces because $f_1 = f_2 = \varepsilon$ and ε is responsible for the accommodation of any gas macro-parameter on the particle surface.

Contributions from the radiation and molecular heat transfer in the denominator (9) for stratospheric and mesospheric aerosols differ greatly; and

$4\sigma e T_\infty^4 / f_2 p_\infty \sqrt{2kT_\infty/\pi m} \ll 1$ for particles with $0.1 < R_p < 5$ μm starting from altitudes of 20–25 km. When ignoring the effect of radiation cooling of a particle for scheme (3), an unexpected result follows from Eq. (9), i.e., the accommodation force is absent at all (due to $f_1 = f_2 = \varepsilon$). However, for scheme (4) we obtain nonzero result of the form

$$F_{\rm acc} = \frac{\pi}{8} R_p^2 \frac{1}{\bar{v}_g} I Q_{\rm abs} \Delta A_n, \qquad (10)$$

where

$$\Delta A_n = \alpha_n(2)/[1-\frac{9\pi}{32}(1-\alpha_n(2))] - \alpha_n(1)/[1-\frac{9\pi}{32}(1-\alpha_n(1))];$$

$\bar{v}_g = \sqrt{8kT_\infty/\pi m}$ is the heat velocity of gas molecules. It can be shown that equation (10), when considering the relation (6), can be reduced to the form (1) obtained in Ref. 11–12 at the same assumptions (free-molecular regime and the neglected radiation cooling of a particle). However, instead of the difference between coefficients of energy accommodation Δa_E, the complex ΔA_n occurs in Eq. (10), which is responsible for accommodation of the normal pulse of gas molecules on different particle hemispheres. The reason of this basic difference can be explained as follows. As indicated in Introduction, the gravito-photophoresis theory in the free molecular regime[11–12] directly borrows results from Ref. 21 on the development of the radiometric method of determination of the energy accommodation coefficients. In review,[33] these results were analyzed in detail and it was shown that in the radiometric Knudsen method, probably, a_n rather than a_E is measured at various temperatures of the heated plate and cold gas. It actually determines the radiometric pressure in the free molecular regime and, therefore, can be called the radiometric accommodation coefficient. Its identification with the energy accommodation coefficient in Ref. 21 took place due to definite simplifying assumptions.[33] Thus, the developed gas-kinetic theory automatically results in the correct form of Eq. (10) and in appearance of ΔA_n in it at a clear physical treatment of the obtained result. A reliable information on values of the normal pulse accommodation coefficients a_n in contrast to the energy accommodation coefficients a_E [Ref. 34] is very scanty and for aerosols it, obviously, is lacking at all. In particular, in Ref. 35 the values of the coefficients a_n given for some gases on the unpurified surfaces of some metals, were measured by the method, which is the development of the Knudsen radiometric method[21] (Table 1).

Table 1. The values of the accommodation coefficients of the normal pulse α_n obtained for unpurified ("technical") metal surfaces[35]

Gas	Wolfram	Tantalum	Germanium
Argon	0.94	0.91	0.92
Helium	0.59	0.61	0.62
Oxygen	0.78	0.80	0.81
Nitrogen	0.76	0.75	0.79

Based on these data, the estimates of ratios of the accommodation force (10) to the gravity and forces of radiometric photophoresis (both solar[5] and thermal[7]) at characteristic values of parameters (Table 2) were obtained.

The estimates show that the accommodation forces may be significant for vertical transfer of stratospheric aerosol, but they do not exceed the gravity. Note that the theory of gravito-photophoresis at identical assumptions and values $\Delta a_E = 0.12 - 0.15$ (the known experimental data for polished and blackened platinum in air) gives the value $F_{\rm gph}/F_{\rm mg} \approx 3$ at altitudes of about 30 km.[17]

The estimate $\Delta\alpha_n = \alpha_n(2) - \alpha_n(1) = 0.05$, possibly, excessive, was chosen as characteristic. Estimates show the accommodation forces to be significant for vertical transport of atmospheric aerosol, but they are lower than the gravity. Note that gravito-photophoresis theory at identical suppositions and $\Delta\alpha E = 0.12 \div 0.15$ (well-known experimental data for polished and blackened surfaces of platinum in air) yields $F_{\rm gph}/F_{\rm mg} \sim 3$ at heights of about 30 km [Ref. 17].

The data[35] do not point to large possible values of Δa_n for surfaces with different material characteristics in air as contrary to rather large possible values of Δa_E for different surfaces in the same gas.[34]

Table 2. Comparison of accommodation force, gravity, and photophoretic forces for carbon particles at a 30 km altitude (R_p = 1 μm, ρ_p = 1 g/cm³, λ_p = 0.12 W/(m · K))

$F_{\rm ac}$ *	$F_{\rm mg}$	$F_{\rm ph}^S$ **	$F_{\rm ph}^{\rm th}$ ***	$F_{\rm acc}/F_{\rm mg}$	$F_{\rm acc}/F_{\rm ph}^S$	$F_{\rm acc}/F_{\rm ph}^{\rm th}$
$1.19 \cdot 10^{-14}$ H	$4.11 \cdot 10^{-14}$ H	$2.40 \cdot 10^{-14}$ H	$0.143 \cdot 10^{-14}$ H	0.29	0.50	1.67
$0.239 \cdot 10^{-14}$ H				0.058		

N o t e s : * The accommodation force $F_{\rm acc}$ is calculated by Eq. (10) at $\alpha_n(2) = 0.80$; $\alpha_n(1) = 0.75$; $Q_{\rm abs} \approx 1$; the intensity of short-wave solar radiation I^S = 1368 W/m² (upper value); intensity of long-wave thermal radiation $I^{\rm th}$ = 275 W/m² (low value). ** The force of solar photophoresis $F_{\rm ph}^{\rm th}$ for carbon particles is calculated at I^S = 1368 W/m²; $N = 1.95 + 0.7i$; λ = 0.5 μm; $J_1 = -0.383$ [Refs. 5, 6]. *** The force of thermal photophoresis $F_{\rm ph}^{\rm th}$ for carbon particles is calculated at $I^{\rm th}$ = 275 W/m²; $N = 2.42 + 1.02i$; λ = 10.6 μm; $J_1 = -0.0989$ [Refs. 6, 7].

Thus, the proposed gas-kinetic theory predicts a possibility of appearance of unusual accommodation forces for particles with an artificial asymmetry of surface characteristics in the radiation field, but does not confirm their high efficiency in vertical transport of stratospheric aerosol.

2. Experimental study of accommodation forces

A comparison of characteristic values of accommodation forces with forces of radiometric photophoresis (Table 2) allows one to hope for a possibility of their direct measurement in some model thermal-physical experiment with macro-particles. Such a method was already used in the study of photophoresis in a rarefied gas[32] and confirmed quantitatively all basic conclusions and predictions of the gas-kinetic theory of this phonemonen.[20]

The used by us experimental setup allowed measurements of small forces affecting the macroscopic substances in a rarefied gas. The setup consisted of a thermal stabilized vacuum cell, pumping system, a system for gas filling and measuring the gas pressure, a radiation source (halogen lamp) with a power meter inside the cell, and a system for measuring forces using the torsion balance.

The particle under study (a sphere with a diameter of 1 cm) with well determined and controlled thermal-physical and surface characteristics is hanged up on a quartz rod on the torsion balance; then the operating cell is pumped out, the radiation intensity is measured, and the gas is filled in up to the required pressure. The force, affecting the particle in the directed radiation field, is recorded by the measuring system, preliminary calibrated. The description of the experimental setup, the range of its operating parameters, the calibration procedure, and estimates of the total measurement error for small forces are given in detail in Ref. 32.

As an object for measuring the accommodation forces, a steel particle in helium was chosen ($R_p = 0.52$ cm; $\rho_p = 7.8$ g/cm^3; $\lambda_p = 14.8$ W/(m$\cdot$K)) at sufficiently low pressures corresponding to the near-free-molecular regime of the gas flow (Kn $\approx 5 \div 15$). Three different experimental sequential situations were realized: a well-polished homogeneous particle; a particle with highly rough frontal and polished rear hemispheres; a rough homogeneous particle in the directed radiation field. The particle was not specially blackened. The gas helium was chosen, because its atoms do not adsorb on the metal surfaces[34]; the system "steel—helium" provided for a fairly large value of the thermal physical parameter $\Lambda = \lambda_p/\lambda_g \approx 100$, at which the photophoretic force is already sufficiently small,[20] that makes it possible to record in the experiment the accompanying accommodation force. Besides, the system "steel—helium" has demonstrated in the experiments with photophoresis[32] the minimal value from the observed values of the radiometric accommodation complex f_1 from Eq. (8). Note that for the system "steel — air" the analogous value was equal to unit, that made impossible to use it in this experiment. Only photophoretic forces act on homogeneous polished and rough particles in the radiation field, and both photophoretic and accommodation forces act on the two-sided asymmetric particle. The measurement of the forces in three considered situations gave the sought information on the magnitude and characteristics of the accommodation force under study.

Figure 2 shows the measurement results on the forces acting on two-sided and homogeneous rough particles, as well as on the difference between these forces, which actually is equal to the accommodation force.

It is evident that a greater measured force corresponds to the rough particle rather than to the two-sided one. It is known that the roughness increases values of coefficients of the energy accommodation and the normal pulse,[34] to which both the photophoretic and accommodation forces are proportional. Undoubtedly, the experimental difference of forces (the accommodation force) is reliably recorded, although its magnitude almost coincides with limiting sensitivity of the experimental setup. It follows from the gas-kinetic analysis that the dependence $F_{ph}(\mathrm{Kn}) = C_1 + C_2/\mathrm{Kn}$ is valid for the near-free-molecular regime of the dimensional photophoretic force.

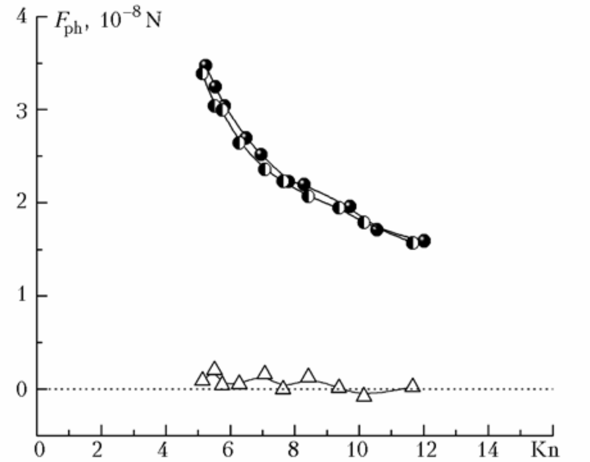

Fig. 2. Results of measurement of forces for the model steel particle in helium. Black small circles denote the homogeneous rough particle; black and open circles denote the two-sided particle with polished and rough hemispheres; triangles denote the difference of forces tested by a rough and two-sided particles; $I = 3000$ W/m^2; $J_1 = -0.28 \pm 0.03$.

Experimental data for homogeneous particles (both for polished and rough) fully support this dependence based on statistical criteria of the assessment. For the two-sided particle this dependence is also statistically reliable, that is indicative of the predominance of photophoretic component in the measured total force.

Finally, the statistical analysis of the force difference has shown that for the dimensional accommodation force the dependence $F_{acc} \propto C/\mathrm{Kn}$ is fulfilled with an acceptable accuracy (Fig. 3).

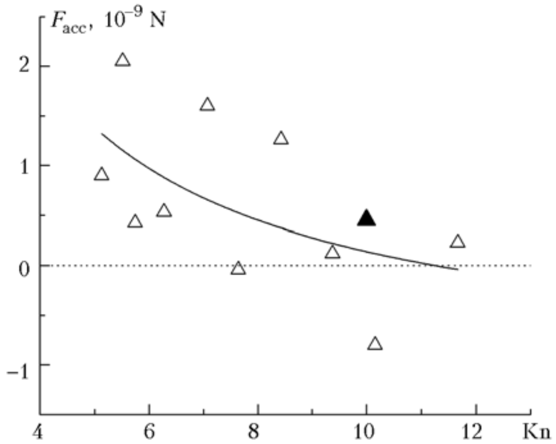

Fig. 3. The difference of forces acting on the homogeneous rough and two-sided steel particles in helium. Open triangles denote the experimental values; black triangle denotes the independent theoretical calculation by Eq. (10) at Kn = 10; the curve denotes the approximation of experimental data by the dependence $F_{acc} = C_1 + C_2/\mathrm{Kn}$ at $C_1 = (-1.11 \pm 0.96) \cdot 10^{-9}$ N, $C_2 = (12.5 \pm 6.6) \cdot 10^{-9}$ N.

The use in the analysis of data for the dimensionless photophoretic force

$$F^*_{\mathrm{ph}} = F_{\mathrm{ph}} / F^{\mathrm{fm}}_{\mathrm{ph}},$$

where

$$F^{\mathrm{fm}}_{\mathrm{ph}} = \frac{\pi}{3} R_{\mathrm{p}}^2 p_\infty I J_1 \Bigg/ \left(p_\infty \sqrt{\frac{2kT_\infty}{\pi m}} + 4\sigma e T_\infty^4 + \frac{\lambda_{\mathrm{p}} T_\infty}{R_{\mathrm{p}}} \right)$$

is the photophoretic force in the free-molecular regime at a full accommodation of the pulse and energy,[20] makes it possible to assess the experimental values of the above-mentioned accommodation complexes. Really, it follows from Ref. 20 that in the near-free-molecular regime

$$F^*_{\mathrm{ph}}(\mathrm{Kn} \gg 1) \approx f_1(\alpha_E, \alpha_n) \left[1 + \frac{4}{15} \frac{1}{\Lambda} f_3(\alpha_E, \alpha_n) \frac{1}{\mathrm{Kn}} \right], \quad (11)$$

where the radiometric accommodation complex

$$f_1 = \alpha_E \alpha_n \Bigg/ \left[1 - \frac{\pi}{32} (9 - \alpha_E)(1 - \alpha_n) \right]$$

was introduced earlier in Eq. (8). Statistical processing of experimental data, using Eq. (11), yields for the polished particle $f_1 = 0.48 \pm 0.01$; for the rough particle $f_1 = 0.52 \pm 0.01$; $f_1 = 0.49 \pm 0.01$ is the value, averaged over the hemisphere for two-sided particle. The increase of f_1 with increasing degree of the surface roughness is one more expected fact.[34]

Note that the division of the Knudsen coefficients a_E and a_n in the complex f_1 in given experiment is impossible in principle. However, independent measurements of a_E can be made in the model experiment on heat transfer from the same heated particle in the same gas (that is planned to realize). For the mirror-diffuse scheme of boundary conditions (3) $f_1 \equiv \varepsilon$ is the coefficient of molecular diffuse reflection. Thus, the change of the roughness degree of the steel particle (from a well-polished surface to a highly rough one) has resulted in the insignificant (0.04) but reliably recorded in experiments increase of the accommodation radiometric complex f_1 (or the coefficient of diffuse reflection ε).

Thereafter, the independent theoretical evaluation of the accommodation force F_{acc} by Eq. (9) and its comparison with the experimental data become possible. Contributions from the radiation and molecular heat transfer are already comparable for model macroscopic particles at low gas pressures (see Eq. (9)). Independent measurements were conducted in order to study the emittance e for polished and rough steel surfaces in vacuum by the methods given in Ref. 32. At $R_{\mathrm{p}} = 0.52$ cm; $e(2) = 0.48 \pm 0.03$; $e(1) = 0.43 \pm 0.03$; $I = 3000$ W/m^2; $p_\infty = 1.38$ Pa (that corresponds to Kn ≈ 10); $T_\infty = 292$ K; $Q_{\mathrm{abs}} = 0.55 \pm 0.03$ [Ref. 32] from Eq. (9) $F_{acc} = 0.45 \cdot 10^{-9}$ N (a black triangle in Fig. 3) was obtained. The evaluation of the force difference by the experimental approximation from Fig. 3 at Kn = 10 gives $f_{acc} = (0.50 \pm 0.17) \cdot 10^{-9}$ N. Thus, the theoretical and experimental data well agree.

Of great interest is the comparison of values of the accommodation and photophoretic forces, measured simultaneously. At Kn = 10 for a particle with a homogeneous rough surface the experimental approximation gives $F_{\mathrm{ph}} = (18.0 \pm 0.2) \cdot 10^{-9}$ N, and the directly measured values of the photophoretic force are $19.6 \cdot 10^{-9}$ N at Kn = 9.72 and $17.0 \cdot 10^{-9}$ N at Kn = 10.55, respectively.

Thus, the unknown ratio of forces in a given experimental situation is 0.023–0.027.

Conclusion

The developed gas-kinetic theory of accommodation forces in the free-molecular regime explains the mechanism of their occurrence from the difference in values of the accommodation coefficients of a normal pulse of gas molecules (not the power accommodation coefficients) on different sides of a model particle. The evaluations with the use of the known values of α_n show that in the stratosphere the accommodation forces are comparable with forces of the radiometric photophoresis, but do not exceed the gravity. These conclusions both qualitatively and quantitatively differ from the conclusions of the semiempirical theory of the gravito-photophoresis developed for explanation of the experiments described in Refs. 11–15.

The measurements conducted in the model thermal-physical experiment with macro-particles have confirmed the existence of accommodation forces, affecting a particle with artificial asymmetry of surface characteristics, together with the forces of radiometric photophoresis. The experimental values for the system "steel particle–helium" are in good agreement with theoretical predictions; in this case the ratio of the accommodation force to the photophoretic force does not exceed 3%.

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