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Enskog-Landau kinetic equation for multicomponent mixture. Analytical calculation of transport coefficients

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Abstract. The Enskog-Landau kinetic equation is considered to describe non-equilibrium processes of a mixture of charged hard spheres. This equation has been obtained in our previous papers by means of the non-equilibrium statistical operator method. The normal solution of this kinetic equation found in the first approximation using the standard Chapman-Enskog method is given. On the basis of the found solution the flows and transport coefficients have been calculated. All transport coefficients for multicomponent mixture of spherical Coulomb particles are presented analytically for the first time. Numerical calculations of thermal conductivity and thermal diffusion coefficient are performed for some specific mixtures of noble gases of high density. We compare the calculations with those ones for point-like neutral and charged particles.

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Construction of kinetic equations for dense gases and plasma is one of the most important problems in the kinetic theory of classical systems. A sequential kinetic theory of dense systems does not exist yet. The Enskog-Landau kinetic equation has been obtained recently in [1] to describe transport processes in non-equilibrium system of charged hard spheres. This equation has its name due to the structure of total collision integral. This integral contains terms of revised Enskog theory, kinetic mean field theory and Landau-like collision integral [2]. The influence of the last term on system behaviours has been of our main interest. This term is caused by the long-range interactions in a system. In particular, it was shown [2] that in the case of small densities and weak interactions the force autocorrelation function and the entire last term in the total collision integral converts to the usual Landau collision integral for a rarefied plasma.

The new kinetic equation and its collision integral are adequate only for systems which can be modelled by charged hard spheres. The great credit for this result is shared by a choice of interparticle interaction potential in additive-like form: hard spheres interaction plus certain "smooth" long-range part (Coulomb interaction in our case). It allowed to avoid divergency at short distances. Unfortunately, a logarithmic divergency at long distances still remains, and to eliminate it one should introduce a cut-off radius of integration (like a Debye one). But unlike the classical Debye formula, in this case we used a modified one, which takes into account particle sizes σ. For the Enskog-Landau kinetic equation [1] a normal solution has been found by means of the standard Chapman-Enskog method. A stress tensor Π and heat flow vector $\mathbf{q}$ have been obtained as well. Expressions for transport coefficients like bulk κ and shear η viscosities and thermal conductivity λ have been derived from structures of Π and $\mathbf{q}$. Numerical calculation of transport coefficients for neutral and once-ionized argon shows a good agreement between the theory and experimental data. In [3,4] these results were generalized to *non-stationary* non-equilibrium process. Whereas to find the normal solution the Chapman-Enskog method [5] is used in [1,2], the much more powerful method of boundary conditions [6] is used in [3,4]. In the limiting case of a stationary non-equilibrium process, the results of [4] completely convert to those of [1]. For hydrodynamic description of fast processes it is better to use the method of boundary conditions [6].

Application of the theory to a multicomponent system was performed step-by-step. The Enskog-Landau kinetic equation for M-component ($M \geqslant 2$) mixture of charged hard spheres has been proposed in [7]. Just the same, the normal solution, flows and transport coefficients have been found by means of the standard Chapman-Enskog method for a two-component system only. New transport coefficients which appear in multicomponent systems are mutual diffusion $D^{\alpha\beta}$ and thermal diffusion D_{T}^{α} coefficients (here α, β are mixture indices). Numerical calculation of the obtained transport coefficients showed a good agreement between the developed theory, experimental data, results of other theories and MD simulations.

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In view of identifying of a normal solution of the kinetic equation for a multicomponent system of charged particles results of [7] are not the first. Namely, it is worthy to note that the normal solution of the kinetic equation for completely ionized plasma is found in [8] using the standard Chapman-Enskog method in the 3rd approximation. But dense systems of finite size particles are consistently considered for the first time in our papers [1–4,7].

In this letter we present our solution of the Enskog-Landau kinetic equation for a multicomponent mixture of charged hard spheres. Similarly to [7], we use the standard Chapman-Enskog method. It is known [5] that the correction expression for one-particle distribution function in the first approximation can be chosen in two different ways. According to the first one described in [9], the correction for one-particle distribution function of α-kind is proportional to $\mathbf{d}_\alpha$ – a diffusion thermodynamic force of α-kind only. The second way is proposed in [5]. In this case the correction is proportional to a certain superposition of diffusion thermodynamic forces of all components of a mixture. It was shown [10] that the second method gives much better results because, unlike the first method, after crossing to linear thermodynamics equations it is important that Onsager's reciprocal relations obey. Nevertheless, in [7] for a two-component system we used the first method because the complication (or generalization) like in [5] is essential for two-component systems in some cases only. Namely, when the density of particle number of some mixture component (or components) is not conserved. Such a situation can be realized in gas mixtures where chemical reactions between components may take place, or in multicomponent mixtures, where transitions between states with different internal degree of freedoms are possible.

Let us consider the Enskog-Landau kinetic equation for multicomponent mixture of charged hard spheres:

$$\left[\frac{\partial}{\partial t}+\mathrm{i}L(1_\alpha)\right] f_1(x_1^\alpha;t)=\sum_{\beta=1}^{M} I_{\mathrm{coll}}\Big(f_1(x_1^\alpha),f_1(x_2^\beta)\Big), \quad (1)$$

here f_1 denotes the one-particle distribution function, $x\equiv\{\mathbf{r};\mathbf{p}\}$ is a set of phase coordinates in a phase space while $\mathbf{r}$ and $\mathbf{p}$ denote the Cartesian coordinate and particle momentum, respectively. Collision integral of this equation has an additive structure

$$I_{\mathrm{coll}}=I_{\mathrm{HS}}^{(0)}+I_{\mathrm{HS}}^{(1)}+I_{\mathrm{MF}}+I_{\mathrm{L}} \quad (2)$$

and each term in (2) is defined like in [7]: the first and second ones are from so-called hard sphere part of interparticle interaction – collision integral of the revised Enskog theory RET:

$$\begin{aligned}
I_{\mathrm{HS}}^{(0)} &= \int \mathrm{d}\mathbf{v}_2^\beta \mathrm{d}\varepsilon \mathrm{d}b\, b|\boldsymbol{g}^{\alpha\beta}| g_2^{\alpha\beta}(\sigma_{\alpha\beta}|n,\beta) \\
&\quad\times\Big(f_1(\mathbf{r}_1^\alpha,\mathbf{v}_1^{\alpha\prime})f_1(\mathbf{r}_2^\beta,\mathbf{v}_2^{\beta\prime})-f_1(\mathbf{r}_1^\alpha,\mathbf{v}_1^\alpha)f_1(\mathbf{r}_2^\beta,\mathbf{v}_2^\beta)\Big), \\
I_{\mathrm{HS}}^{(1)} &= \sigma_{\alpha\beta}^3\int \mathrm{d}\hat{\mathbf{r}}_{12}^{\alpha\beta}\mathrm{d}\mathbf{v}_2^\beta\theta(\hat{\mathbf{r}}_{12}^{\alpha\beta}\cdot\boldsymbol{g}^{\alpha\beta}) \\
&\quad\times\Big[\hat{\mathbf{r}}_{12}^{\alpha\beta} g_2^{\alpha\beta}(\mathbf{r}_{12}^{\alpha\beta}|n,\beta)\Big(f_1(\mathbf{r}_1^\alpha,\mathbf{v}_1^{\alpha\prime})\boldsymbol{\nabla}f_1(\mathbf{r}_2^\beta,\mathbf{v}_2^{\beta\prime}) \\
&\quad -f_1(\mathbf{r}_1^\alpha,\mathbf{v}_1^\alpha)\boldsymbol{\nabla}f_1(\mathbf{r}_2^\beta,\mathbf{v}_2^\beta)\Big)+\frac{1}{2}\Big(\hat{\mathbf{r}}_{12}^{\alpha\beta}\cdot\boldsymbol{\nabla}g_2^{\alpha\beta}(\mathbf{r}_{12}^{\alpha\beta}|n,\beta)\Big) \\
&\quad\times\Big(f_1(\mathbf{r}_1^\alpha,\mathbf{v}_1^{\alpha\prime})f_1(\mathbf{r}_2^\beta,\mathbf{v}_2^{\beta\prime})+f_1(\mathbf{r}_1^\alpha,\mathbf{v}_1^\alpha)f_1(\mathbf{r}_2^\beta,\mathbf{v}_2^\beta)\Big)\Big];
\end{aligned}$$

the third one is caused by taking into account of long-range interparticle interaction in the mean field approximation KMFT (this term is of the first order in interaction):

$$\begin{aligned}
I_{\mathrm{MF}} &= \frac{1}{m_\alpha}\int \mathrm{d}\mathbf{r}_2^\beta\frac{\partial}{\partial\mathbf{r}_1^\alpha}\Phi^1(|\mathbf{r}_{12}^{\alpha\beta}|) \\
&\quad\times g_2^{\alpha\beta}(\mathbf{r}_1^\alpha,\mathbf{r}_2^\beta;t)n_1(\mathbf{r}_2^\beta;t)\frac{\partial}{\partial\mathbf{v}_1^\alpha}f_1(x_1^\alpha;t);
\end{aligned}$$

and, finally, the third one is the so-called Landau-like collision term (it is of the second order in interaction):

$$\begin{aligned}
I_{\mathrm{L}} &= \frac{1}{4(m^*)^2}\frac{\partial}{\partial\mathbf{v}_1^\alpha}\int \mathrm{d}x_2\; g_2(\mathbf{r}_1^\alpha,\mathbf{r}_2^\beta;t)\left[\frac{\partial\Phi^1(|\mathbf{r}_{12}^{\alpha\beta}|)}{\partial\mathbf{r}_{12}^{\alpha\beta}}\right] \\
&\quad\times\int_{-\infty}^{0}\mathrm{d}t'\left[\frac{\partial\Phi^1(|\mathbf{r}_{12}^{\alpha\beta}+\boldsymbol{g}t'|)}{\partial\mathbf{r}_{12}^{\alpha\beta}}\right]\left[\frac{\partial}{\partial\mathbf{v}_1^\alpha}-\frac{\partial}{\partial\mathbf{v}_2^\beta}\right] \\
&\quad\times f_1(x_1^\alpha;t)f_1(x_2^\beta;t).
\end{aligned}$$

In expressions for $I_{\mathrm{HS}}^{(0)}$, $I_{\mathrm{HS}}^{(1)}$, I_{MF} and I_{L} we used the following designations: b, impact parameter; β, an analogue of local inversed temperature; ε, azimuthal angle of scattering; $\boldsymbol{g}^{\alpha\beta}$, relative velocity of α- and β-kind particles; $g_2^{\alpha\beta}$, two-particle correlation function; m^*, reduced mass; m_α, partial masses of particles; n, total density of particles number; n_α, partial densities of particles numbers; $\hat{\mathbf{r}}_{12}^{\alpha\beta}$, unit vector along $\mathbf{r}_{12}^{\alpha\beta}$ direction; $\mathbf{v}'$, velocities of hard spheres after collision; $\theta(\dots)$, Heaviside unit step function.

In the limit of a system of point-like charged particles of low density, the collision integral (2) transforms to the usual Landau collision integral [11,12]. Following a concept of consistent description of kinetics and hydrodynamics of non-equilibrium processes [13–17], kinetic equation (1) should be solved together with local conservation laws [4] for additive invariants. These additive invariants in collisions (or scattering) of charged hard spheres are mass (or total density), momentum and total energy [5,9,11,12]. It should be noted that for rarefied systems it was sufficient to consider kinetic energy only, while in dense systems potential interaction energy is essential and cannot be neglected.

The solution of equation (1) found in the first approximation by means of the Chapman-Enskog method is

$$f_1^{(1)}(x_1^\alpha;t) = f_1^{(0)}(x_1^\alpha;t)\Big[1+\varphi(x_1^\alpha;t)\Big], \tag{3}$$

where $f_1^{(0)}(x_1^\alpha;t)$ is the local quasi-equilibrium Maxwell one-particle distribution function:

$$f_1^{(0)}(x_1^\alpha;t) = n_\alpha(\mathbf{r}_1;t)\left[\frac{m_\alpha}{2\pi k_\mathrm{B}T(\mathbf{r}_1;t)}\right]^{3/2}\exp\left\{-\frac{m_\alpha\left(c_1^\alpha(\mathbf{r}_1;t)\right)^2}{2k_\mathrm{B}T(\mathbf{r}_1;t)}\right\}.$$

This function is the solution of equation (1) in the zeroth approximation and satisfies the Fredgolm conditions. Correction $\varphi(x_1^\alpha;t)$ reads:

$$\begin{aligned}\varphi(x_1^\alpha;t) = &-A_\alpha[(C_1^\alpha)^2]\mathbf{C}_1^\alpha(\mathbf{r}_1;t)\cdot\boldsymbol{\nabla}\ln T(\mathbf{r}_1;t)\\ &-B_\alpha[(C_1^\alpha)^2]\left[\mathbf{C}_1^\alpha\mathbf{C}_1^\alpha-\frac{1}{3}(C_1^\alpha)^2\mathsf{I}\right]:\boldsymbol{\nabla}\mathbf{V}(\mathbf{r}_1;t)\\ &+n\sum_{\beta=1}^{M}E_{\alpha\beta}[(C_1^\alpha)^2]\mathbf{C}_1^\alpha(\mathbf{r}_1^\alpha;t)\cdot\mathbf{d}_\beta(\mathbf{r}_1;t).\end{aligned} \tag{4}$$

Here

$$\mathbf{C}^\alpha=\mathbf{C}^\alpha(\mathbf{r};t)=\left[\frac{m_\alpha}{2k_\mathrm{B}T}\right]^{1/2}\mathbf{c}^\alpha(\mathbf{r};t),\ \mathbf{c}^\alpha(\mathbf{r};t)=\mathbf{v}^\alpha-\langle\mathbf{v}\rangle.$$

In other words, $\langle\mathbf{v}\rangle$ is nothing but hydrodynamical velocity $\mathbf{V}(\mathbf{r};t)$. Functionals $A_\alpha[(C_1^\alpha)^2]$, $B_\alpha[(C_1^\alpha)^2]$, $E_{\alpha\beta}[(C_1^\alpha)^2]$ are defined by the Sonine-Laguerre polynomials [5,9].

Having the solution of equation (1) in the first approximation, one can calculate the stress tensor Π, heat flow vector $\mathbf{q}$ and diffusion velocity $\mathbf{V}^\mathrm{d}$ in the same approximation. The expression for Π reads:

$$\Pi = p\mathsf{I}-\kappa\left(\boldsymbol{\nabla}\cdot\mathbf{V}\right)\mathsf{I}-2\eta\mathsf{S},$$

where p is total pressure in the first approximation (I is the unit tensor, S is the velocities shift tensor):

$$\begin{aligned}p = &\,nk_\mathrm{B}T+\frac{2}{3}\pi k_\mathrm{B}T\sum_{\alpha,\beta=1}^{M}\sigma_{\alpha\beta}^3 g_2^{\alpha\beta}n_\alpha n_\beta\\ &-\frac{2}{3}\pi\sum_{\alpha,\beta=1}^{M}n_\alpha n_\beta\int_{\sigma_{\alpha\beta}}^{\infty}\mathrm{d}x\,x^3 g_2^{\alpha\beta}(x)\frac{\partial}{\partial x}\Phi_{\alpha\beta}^\mathrm{l}(x),\end{aligned}$$

($g_2^{\alpha\beta}(x)$ is the binary quasi-equilibrium correlation function, $\Phi_{\alpha\beta}^\mathrm{l}(x)$ is a long-range potential of interaction); κ is the bulk viscosity of a multicomponent mixture:

$$\kappa=\frac{4}{9}\sum_{\alpha,\beta=1}^{M}\sigma_{\alpha\beta}^4 g_2^{\alpha\beta}n_\alpha n_\beta\sqrt{2\pi k_\mathrm{B}T\mu_{\alpha\beta}}=\sum_{\alpha,\beta=1}^{M}\kappa_{\alpha\beta}, \tag{5}$$

η is the shear viscosity of a multicomponent mixture:

$$\begin{aligned}\eta = &\frac{3}{5}\kappa+\frac{1}{2}k_\mathrm{B}T\sum_{\alpha=1}^{M}n_\alpha B_\alpha(0)\\ &+\frac{2}{15}\pi k_\mathrm{B}T\sum_{\alpha,\beta=1}^{M}\sigma_{\alpha\beta}^3 g_2^{\alpha\beta}\mu_{\alpha\beta}n_\alpha n_\beta\left[\frac{B_\alpha(0)}{m_\alpha}+\frac{B_\beta(0)}{m_\beta}\right].\end{aligned} \tag{6}$$

The expression for heat flow $\mathbf{q}$ reads:

$$\mathbf{q}=-\lambda\boldsymbol{\nabla}T+\sum_{\alpha=1}^{M}\omega_\alpha\mathbf{d}_\alpha.$$

Quantities ω_α are connected with a matter transfer due to a temperature gradient (the Soret effect) and due to a heat transfer caused by a gradient of concentration (the Dufour effect). If one also takes into account a barrodiffusion process, this constitutes the total contribution into a heat flow from cross transfer processes. λ is the thermal conductivity coefficient of a multicomponent mixture:

$$\begin{aligned}\lambda = &\sum_{\alpha,\beta=1}^{M}\frac{3k_\mathrm{B}\,\kappa_{\alpha\beta}}{m_\alpha+m_\beta}-\sqrt{2k_\mathrm{B}^3T}\\ &\times\left[\frac{5}{4}\sum_{\alpha=1}^{M}\frac{n_\alpha}{\sqrt{m_\alpha}}\left[A_\alpha(1)-A_\alpha(0)\right]\right.\\ &\left.+\frac{2\pi}{3}\sum_{\alpha,\beta=1}^{M}\frac{\sigma_{\alpha\beta}^3 g_2^{\alpha\beta}n_\alpha n_\beta}{m_\alpha+m_\beta}\left[\frac{3\mu_{\alpha\beta}}{\sqrt{m_\beta}}A_\beta(1)-\sqrt{m_\beta}A_\beta(0)\right]\right].\end{aligned} \tag{7}$$

It should be noted, however, that not λ, but the heat conductivity coefficient χ is measured experimentally. It is mutually connected with λ by the relation $\chi=\lambda/(\rho C_p)$, where C_p is the heat capacity at constant pressure. Diffusion velocity in the first approximation reads:

$$\mathbf{V}_\alpha^\mathrm{d}=-D_\mathrm{T}^\alpha\boldsymbol{\nabla}\ln T-\sum_{\beta=1}^{M}D^{\alpha\beta}\mathbf{d}_\beta,$$

where

$$D_\mathrm{T}^\alpha=\sqrt{\frac{k_\mathrm{B}T}{2m_\alpha}}A_\alpha(0) \tag{8}$$

is the thermal diffusion coefficient of a mixture, while

$$D^{\alpha\beta}=-n\sqrt{\frac{k_\mathrm{B}T}{2m_\alpha}}E_{\alpha\beta}(0) \tag{9}$$

is the mutual diffusion coefficient. Quantities $B_\alpha(0)$ in (6), $A_\alpha(0)$ and $A_\alpha(1)$ in (7) and (8), $E_{\alpha\beta}(0)$ in (9) are nothing but coefficients of expansion in the Sonine-Laguerre polynomials. Their general definition for an arbitrary potential of interaction is given in [5]. The calculations for a special case of a two-component mixture is performed in [7]. All the obtained quantities $B_\alpha(0)$, $A_\alpha(0)$, $A_\alpha(1)$ and $E_{\alpha\beta}(0)$ ultimately depend on the so-called Ω-integrals.

Numerical calculation for transport coefficients κ, η, λ, D_T^α and $D^{\alpha\beta}$ has been performed. For two- and three-component mixtures of neutral and charged hard spheres we studied the dependences of transport coefficients on density, temperature, and concentration ratio of some mixture components [18]. There are a lot of approaches which allow to calculate viscosity with sufficient accuracy. However, these approaches do not allow to calculate well thermal conductivity for dense

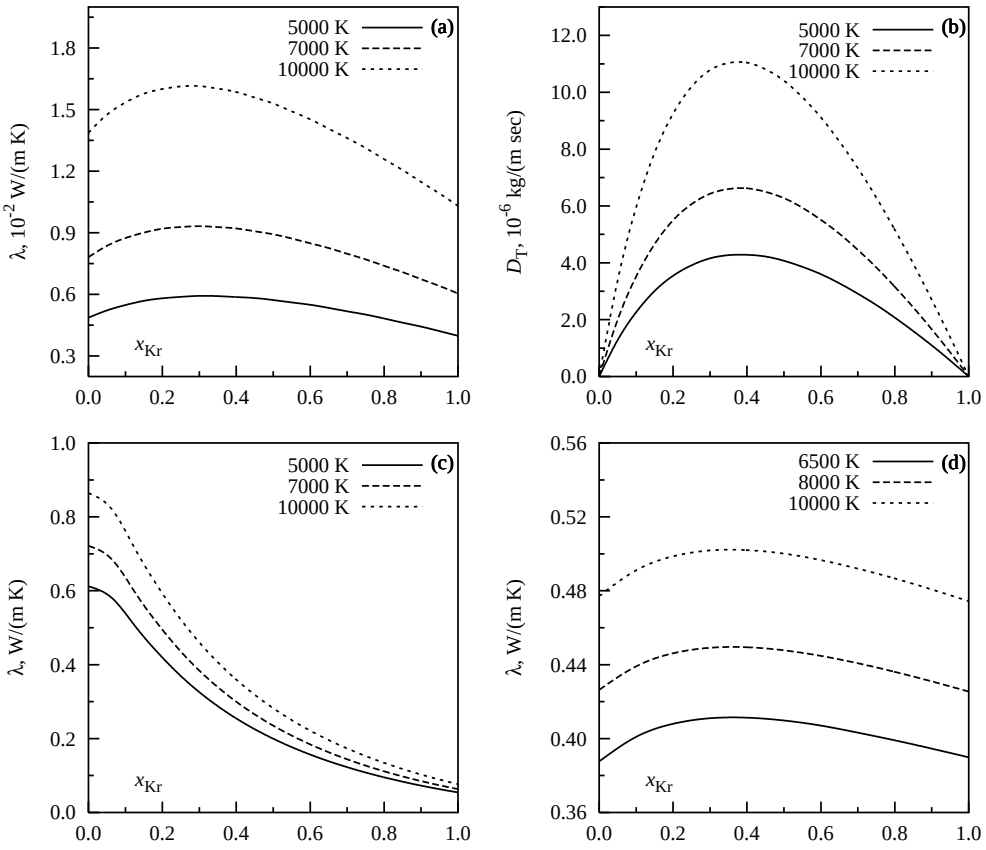

Fig. 1. Concentration ratio dependences of transport coefficients of some binary and ternary systems of neutral and charged hard spheres. Thermal conductivity (a) and thermal diffusion coefficient (b) of a system Ar^+–Kr^+ at total concentration $n = 2 \times 10^{20}$ cm^{-3}. Thermal conductivity of a system He–Kr–Ar^+ (c) at total concentration $n = 2 \times 10^{21}$ cm^{-3}, $x_{Ar} = 0.2$ or $n_{Ar} = 4 \times 10^{20}$ cm^{-3}. Thermal conductivity of a system He–Ar^+–Kr^+ (d) at total concentration $n = 1.25 \times 10^{21}$ cm^{-3}, $x_{He} = 0.6$.

and moderately dense systems. Our theory is devoid of such a circumstance. This is reached due to the following: firstly, the Enskog-Landau kinetic equation is obtained by means of the non-equilibrium statistical operator method from the first principles of statistical mechanics without phenomenological assumptions; secondly, kinetics and hydrodynamics in the studied systems are considered simultaneously. The last factor is very important for dense and moderately dense systems [13–17]. As a result, we obtain a good agreement between the theory and experimental data. In Figure 1 we present the concentration ratio dependences of some transport coefficients for two- and three-component mixtures for charged hard spheres. In all cases, the total concentration is considered to be constant. Figure 1b illustrates the limiting cases for the thermal diffusion coefficient D_T^α of a system Ar^+–Kr^+ when $x_{Kr} \to 0$ or $x_{Kr} \to 1$, that is for one-component systems. In this case, diffusion thermodynamic forces vanish and thermal diffusion vanishes too. Figure 2 shows numerical calculations for the same systems as in Figure 1, but for point-like neutral and charged particles. Transport coefficients for last ones were calculated as for the usual Boltzmann-Landau kinetic equation [11]. The magnitude of transport coefficients therewith slows down. It is interesting to note that in three-component systems of point-like particles (Figs. 2c and d) transport coefficients depend on Λ slightly. It is well to bear in mind that this takes place if at least one component is not charged.

From the present letter, one can draw the following conclusions. The obtained Enskog-Landau kinetic equation for charged hard spheres turned out to be very useful for several purposes. First of all, the collision integral of this equation does not contain a divergency at small distances. Secondly, the normal solution and *all* transport coefficients have analytical structure. They can be easily used to study some specific systems. Finally, the analytical structure of transport coefficients allows us to find fast and easily systems, which can be best described by the obtained kinetic equation, as well as density and temperature ranges, where the agreement between the theory and experimental data is the closest. At the same time, our theory has not met with success in treatment of the free electrons role. Basing on the conclusions by Ichimaru *et al.* [19], consistent treatment of electrons is possible only within the frame of quantum kinetic theory. Our theory

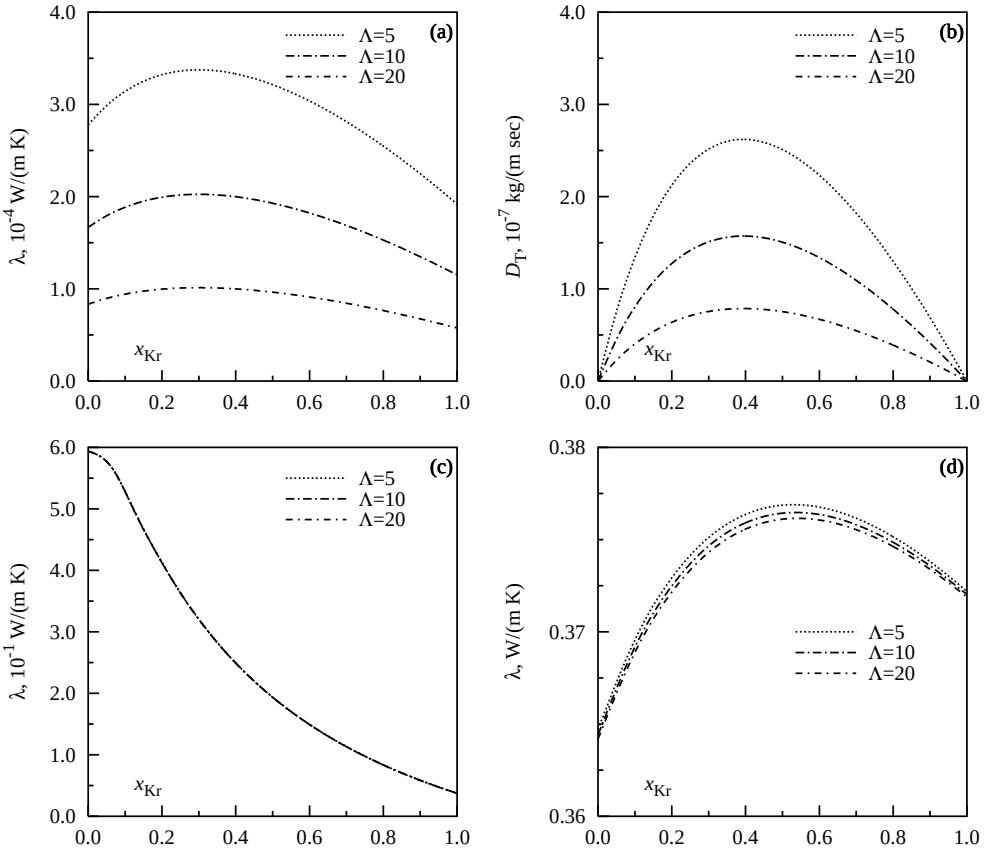

Fig. 2. The same as in Figure 1, but for systems of neutral and charged point-like particles at $T = 5000$ K for (a), (b) and (c) and at $T = 6500$ K for (d). Parameter Λ indicates for what value of Coulomb logarithm calculation was performed.

is purely classical. Just the same, our papers [1,7] show very good agreement between theoretical calculations and experimental data.

The next step in this theory is to calculate a dynamical screening radius in a system. Partially this problem has been already solved in our recent paper [20].

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