Transport phenomena in non-uniform gas subjected to laser radiation

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Abstract. The paper discusses the theory of transport processes in one-component gas located in capillary subjected to resonant laser radiation and both temperature and pressure gradients. The equations for the kinetic coefficients determining heat- and mass transport in the gas are derived on the basis of modified Boltzmann equations for the excited and unexcited particles. The cross kinetic coefficients satisfy the Onsager reciprocity for all Knudsen numbers and laws of gas particles interaction with each other and with boundary surface of the capillary. Analysis of possible non-equilibrium stationary states of first and second order for the one-component gas in the capillary has been developed on the basis of the Prigogine theorem of stationary states. Equations describing the stationary states in Knudsen limit ($Kn \gg 1$) and slip-flow regime ($Kn \ll 1$) were derived.

1. Introduction

Thermodynamic description of transport phenomena is based on the laws of conservation of mass, momentum and energy and the principle of increasing entropy. According to the thermodynamics of irreversible processes[1] the entropy production in the system is determined as a sum of products of generalized fluxes and generalized forces bound to those fluxes. These forces are generally associated with spatial inhomogeneity of thermodynamic parameters of the system or with a deviation of some of the internal variables from their equilibrium values [1].

Specific feature of light-induced transport phenomena is that they take place at homogeneous thermodynamic parameters of gas and under certain conditions and can generate temperature and pressure gradients [2]. These light-induced gradients stimulate their own fluxes of mass and heat in the gas. Theoretical description of these phenomena at the phenomenological level is impossible since the mechanisms of light-induced transport phenomena are associated only with the molecular-kinetic properties of the gas. Development of Onsager kinetic theory of these processes is the main objective of this study.

Laser radiation under resonant interaction with the gaseous medium behaves as the «two-faced Janus». The radiation can simultaneously violate the gas equilibrium by internal and translational degrees of freedom. This means that radiation stimulates two types of fluxes, «scalar» flux, characterizing change of population of the particles energy levels compared to their equilibrium values and vector fluxes of mass and heat of the gas.

Disequilibrium due to translational degrees of freedom is the result of asymmetry in the distribution functions of the excited and unexcited particles related to zero- component of the particles velocity vector in the direction of radiation propagation (Figure 1).
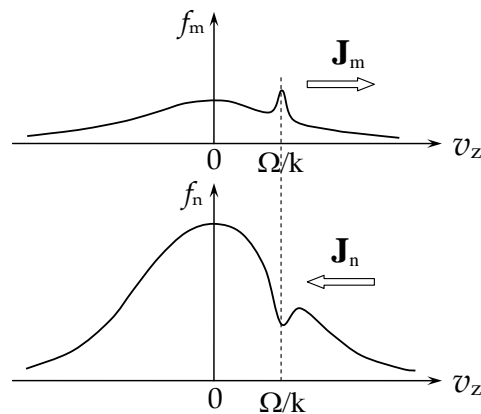
Figure 1. The distribution functions of excited and unexcited particles.

In accordance with the Doppler effect, radiation interacts only with those particles having Doppler shift **kv** (**k** is the wave vector, **v** is the velocity vector of the particle) compensates detuning Ω of radiation frequency ω relatively to ω_{mn}, the $m - n$ frequency transition from the ground state of particle n to excited state m. In linear approximation, when the magnitude of the particle velocity is much less than light speed, Doppler shift is determined by the expression

$$\mathbf{kv} = \Omega = \omega - \omega_{mn} \,, \qquad |\Omega| << \omega, \omega_{mn} \tag{1}$$

As a result of light absorption, distributions of excited and unexcited particles near the resonant velocity $v_p = \Omega/k$ are distorted due to the appearance of Bennett's peak and dip [3] (Figure 1). The asymmetry of velocity distributions at $\Omega \neq 0$ means the existence of fluxes of excited $\mathbf{J}_m$ and unexcited $\mathbf{J}_n$ particles having opposite directions. The particles absorbing light change their transport characteristics. If buffer gas [2, 4] or boundary surface with which m- and n-particles interact differently [5-7] are available in the system, these fluxes do not prevent each other and gas as a whole moves. The flux of the absorbing gas $\mathbf{J} = \mathbf{J}_n + \mathbf{J}_m$ is known as the light-induced drift – LID [4-7]. Simultaneously with LID, resonant laser radiation stimulates light-induced heat transfer in the gas, designated as LIHT [8].

The problem becomes much more complicated if the state of the gas is governed by the pressure and temperature gradients in addition to the resonant radiation. Each of these gradients creates its own fluxes, interacting with the light-induced mass and heat fluxes. In the closed gaseous systems under certain conditions this interaction leads to occurrence of various nonequilibrium steady states.

Cross-transport processes in one-component gas in the presence of laser radiation and both pressure and temperature gradients were overviewed in [9]. The paper was focused on the discussion of light-induced baro- and thermo- effects that take place in closed heat insulated systems. However the proof of an Onsager reciprocity for the cross kinetic coefficients was not given in this reference. In addition, the entropy production due to the interaction of gas particles with the surface of the capillary was only stated, but not analyzed.

The objectives of this work are as follows.

First, based on the kinetic theory of gases we derive the expression for the entropy production in one-component gas in the field of the laser radiation in presence of the pressure and temperature gradients; and determine the generalized thermodynamic forces and corresponding fluxes;

Second, we prove Onsager's reciprocity for the cross kinetic coefficients at arbitrary Knudsen numbers and any nature of gas particles interaction with each other and boundary surface.

Third, using theorems of the non-equilibrium thermodynamics we consider the possible stationary states of the gas.

2. Statement of the problem

We consider one-component gas in the capillary. Let capillary length L is much greater than its radius r_0 (L $>> r_0$). The resonant optical radiation is uniform over the cross section of the capillary and propagates along the axis of the capillary as a traveling monochromatic wave of ω frequency. In addition, the state of the gas is disturbed by the longitudinal temperature and pressure gradients.

We will use the two-level approximation according to which the particles can be either in the ground state *n*, or in the excited state *m*. Particles absorbing radiation transit to an excited state. They change their transport properties, in particular, the collision cross-sections and accommodative characteristics. The process of radiative decay of the excited *m* level with frequency Γ_m occurs simultaneously with the induced transitions. Thus, the gas can be considered as a mixture consisting of particles of equal mass m_0 but with different interaction cross-sections and accommodative properties. The components continuously exchange the particles.

The state of gas is described by distribution functions of excited f_m and unexcited f_n particles that satisfy the Boltzmann kinetic equations including terms responsible for the interaction of particles with radiation [10]:

$$\frac{\partial f_i}{\partial t} + \mathbf{v}\nabla f_i = a_i R + I_i ,$$ (2)

$$R = \frac{\Gamma_m \chi(\mathbf{v})}{2}(f_m - f_n) + \Gamma_m f_m , \qquad \chi(\mathbf{v}) = \frac{4|G_{mn}|^2 \Gamma}{\Gamma_m \left[\Gamma^2 + (\Omega - \mathbf{kv})^2\right]} , \qquad G_{mn} = \frac{E d_{mn}}{2\hbar} ,$$

$$a_i = \begin{cases} 1, & i = n \\ -1, & i = m \end{cases} , \qquad I_i = I_{ii} + I_{ij} , \qquad i, j = n, m .$$

Here Γ is homogeneous half-width of the absorption line, *E* is the electric field amplitude of the light wave, d_{mn} is the dipole moment of *m–n* transition, **k** is the wave vector, and I_{ij} are Boltzmann integrals of collisions between the particles of *i*-th and *j*-th kind. Rabi frequency G_{mn} determines the rate of induced transitions between the levels of particles per unit time and depends on the intensity of radiation. The saturation parameter $\chi(\mathbf{v})$ characterizes the probability of radiation absorption per unit time by the particles which are moving with speed **v** and determines the width of the Bennett's peak.

The boundary conditions for the distribution functions of particles have the form [11]:

$$|\mathbf{v}\cdot\mathbf{n}| f_i(\mathbf{v}) = \sum_{j=n,m} \int_{(\mathbf{v'n})<0} B(j, \mathbf{v'} \to i, \mathbf{v}) f_j(\mathbf{v'}) |\mathbf{v'}\cdot\mathbf{n}| d\mathbf{v'}$$ (3)

$$(\mathbf{v}\cdot\mathbf{n}) > 0 \qquad (i, j) = m, n ,$$

where **n** is inward normal to the surface of the capillary, $f_i(\mathbf{v})$ and $f_i(\mathbf{v'})$ are distribution functions of the reflected and incident particles on the wall of *i*-th kind, respectively, $B(j, \mathbf{v'} \to i, \mathbf{v})$ is the scattering kernel (the probability density is such that the internal energy E_j of the state *j* and the velocity $\mathbf{v'}$ of the incident particle on the wall after reflection is equal to E_i and **v**). The scattering kernel satisfies the following conditions [11]:

normalization

$$\sum_{j=n,m} \int_{(\mathbf{vn})>0} B\left(j, \mathbf{v}' \to i, \mathbf{v}\right) d\mathbf{v} = 1 \tag{4}$$

reciprocity relation expressing the principle of detailed balance on the gas—surface border

$$\left|\mathbf{v}' \cdot \mathbf{n}\right| f_{j0}\left(\mathbf{v}'\right) G_j B\left(j, \mathbf{v}' \to i, \mathbf{v}\right) = \left|\mathbf{v} \cdot \mathbf{n}\right| f_{i0}\left(\mathbf{v}'\right) G_i B\left(i, -\mathbf{v} \to j, -\mathbf{v}'\right) \tag{5}$$

where f_{i0} is the equilibrium Maxwell-Boltzmann distribution function, G_i is the degree of degeneracy.

Integrating (5) over velocity space $\left(\mathbf{v}' \cdot \mathbf{n}\right) < 0$ and summing over the states j, taking into account the normalization (4), we obtain

$$\left|\mathbf{v} \cdot \mathbf{n}\right| f_{i0}\left(\mathbf{v}\right) = \sum_{j=n,m} \int_{(\mathbf{v'n})<0} B\left(j, \mathbf{v}' \to i, \mathbf{v}\right) f_{j0}\left(\mathbf{v}'\right) \left|\mathbf{v}' \cdot \mathbf{n}\right| d\mathbf{v}' \tag{6}$$

We choose a cylindrical coordinate system (r, ϕ, z), so that the polar axis z is directed along the axis of the capillary.

In the weak-field approximation ($\chi(\mathbf{v}) \ll 1$) and at small pressure and temperature gradients, the distribution functions of excited and unexcited particles are insignificantly different from local equilibrium Maxwell-Boltzmann distributions:

$$f_i = f_{i0}\left[1 + h_i\left(\mathbf{r}, \mathbf{v}\right)\right] \tag{7}$$

The local equilibrium Maxwell-Boltzmann distribution looks like

$$f_{i0} = n_i\left(z\right)\left(\frac{m_0}{2\pi k_B T\left(z\right)}\right)^{3/2} \exp\left(-\frac{m_0 v^2}{2 k_B T\left(z\right)}\right),$$

$$n_i\left(z\right) = n\left(z\right) \frac{\exp\left(-E_i/k_B T\left(z\right)\right)}{\sum_l \exp\left(-E_l/k_B T\left(z\right)\right)}. \tag{8}$$

Here $T\left(z\right), n\left(z\right), n_i\left(z\right)$ are the local values of the gas temperature, the total particle number density, and the population of the i-th level, respectively; k_B is the Boltzmann constant.

Due to linearization (7) and equation (6), the boundary conditions (3) on the wall of the capillary for the perturbation functions take the following form:

$$\left|\mathbf{v} \cdot \mathbf{n}\right| f_{i0} h_i\left(\mathbf{v}\right) = \sum_{j=n,m} \int_{(\mathbf{v'n})<0} B\left(j, \mathbf{v}' \to i, \mathbf{v}\right) f_{j0} h_j\left(\mathbf{v}'\right) \left|\mathbf{v}' \cdot \mathbf{n}\right| d\mathbf{v}' \tag{9}$$

To make a separate analysis for vector fluxes directed along the capillary and scalar fluxes associated only with the change of the level population we distinguish even and odd parts of the longitudinal component of the particles v_z velocity vector in perturbation functions and in saturation parameter

$$\chi(v_z) = \chi^+(v_z) + \chi^-(v_z), \qquad h_i(v_z) = h_i^+(v_z) + h_i^-(v_z), \tag{10}$$

$$\chi^{\mp}(v_z) = \frac{\chi(v_z) \mp \chi(-v_z)}{2}, \qquad h_i^{\mp}(v_z) = \frac{h_i(v_z) \mp h_i(-v_z)}{2}.$$

Based on the assumptions made the kinetic equations (2) are linearized with respect to perturbation functions h_i and for steady gas state are written as:

$$\mathbf{v}_\perp \frac{\partial h_i^\mp}{\partial \mathbf{r}} = \sum_{j=n,m} L_{ij}\left(h_i^\mp\right) + b_i h_m^\mp + g_i^\mp \tag{11}$$

where the following notations are used:

$$g_i^- = -v_z \frac{1}{p}\frac{\partial p}{\partial z} - v_z\left(\frac{m_0 v^2}{2kT} - \frac{5}{2} + \varepsilon_i - \bar{\varepsilon}\right)\frac{1}{T}\frac{\partial T}{\partial z} + \frac{\Gamma_m}{2}\chi^-(v_z)\left(\frac{n_{j0}}{n_{i0}} - 1\right),$$

$$g_i^+ = \frac{\Gamma_m}{2}\chi^+(v_z)\left(\frac{n_{j0}}{n_{i0}} - 1\right) + b_i, \qquad v^2 = v_z^2 + v_\perp^2 \qquad (i,j) = n,m \qquad i \neq j \tag{12}$$

$$b_i = \begin{cases} \Gamma_m \dfrac{n_{m0}}{n_{n0}}, & i = n \\ -\Gamma_m, & i = m \end{cases}, \qquad \bar{\varepsilon} = \frac{\sum\limits_l \varepsilon_l \exp(-\varepsilon_l)}{\sum\limits_l \exp(-\varepsilon_l)} = \frac{\bar{E}}{k_B T_0}, \qquad v_\perp^2 = v_r^2 + v_\phi^2, \qquad \varepsilon_i = \frac{E_i}{k_B T_0}.$$

Here $\mathbf{r}$ and $\mathbf{v}_\perp$ are two-dimensional radius vector and vector component of the velocity of particles in the cross-section of the capillary, $L_{ij}(h_i)$ are the linearized collision integrals for particles of i-th and j-th kinds, n_{n0}, n_{m0} are the equilibrium populations of the ground and excited states at the equilibrium temperature of the gas T_0, $\bar{E}$ is the average internal energy of the particles in the equilibrium state. One can see [12] that the linearized collision integral $L_{ij}(h_i)$ off the odd function h_i over velocity v_z is an odd function, i.e. $L_{ij}(h_i(-v_z)) = -L_{ij}(h_i(v_z))$.

The linearized kinetic equations (11) are separated into odd ("minus") and even ("plus") relatively velocity v_z. The first equations describe vector fluxes that are associated with transfer of mass, heat and entropy of the gas along the capillary, second – with the scalar fluxes describing the pressure anisotropy and radiation-collisional heating or cooling gas [10].

3. The equation of entropy balance

Consider a volume V of gas contained in the capillary tube of length *L*. We define its entropy in conventional manner [1]:

$$S_e = -k_B \sum_{i=m,n} \int_V dV \int f_i \ln f_i \, d\mathbf{v} \tag{13}$$

The rate of entropy change in the respective volume of gas is equal to:

$$\frac{\partial S_e}{\partial t} = -k_B \sum_i \int_V dV \int (\ln f_i + 1)\frac{\partial f_i}{\partial t} \, d\mathbf{v} \tag{14}$$

Taking into account the kinetic equation (2) we obtain the entropy balance equation in the form:

$$\frac{\partial S_e}{\partial t} + \int_V div\mathbf{j}\, dV = P_R + P_c \tag{15}$$

where $\mathbf{j}$ is vector flux density of entropy:

$$\mathbf{j} = -k_B \sum_i \int \mathbf{v} f_i \ln f_i d\mathbf{v} \qquad (16)$$

P_R is the total entropy production in the considered volume due to radiation interaction with the gas:

$$P_R = -k_B \int dV \int R \ln\left(\frac{f_n}{f_m}\right) d\mathbf{v} \qquad (17)$$

P_c is the total entropy production due to the interaction of gas particles with each other:

$$P_c = -k_B \sum_i \int dV \int I_i \ln f_i d\mathbf{v} \qquad (18)$$

Consider the stationary state of the system when the entropy of the gas volume does not change in time. In this case, the entropy balance equation (15) can be written as:

$$\int div\mathbf{j}\, dV = P_R + P_c \qquad (19)$$

Using linearization of distribution functions (7) we obtain the entropy balance equation for stationary weakly nonequilibrium state of the system.

Taking into account (11) and (12) the equation (19) is divided into equations with vector («-») and scalar («+») fluxes:

$$\int \left(div\mathbf{j}\right)^{\mp} dV = P_R^{\mp} + P_c^{\mp} \qquad (20)$$

We consider only vector fluxes. To simplify the equations hereafter we omit the superscript «-» in all the physical quantities, believing that function $h_i(v_z)$ is odd relatively to velocity v_z.

Using the Gauss theorem and decomposition $\ln\left(1+h_i\right) \approx h_i - h_i^2/2$, we get:

$$\int div\mathbf{j}\, dV = -k_B \sum_i \int dV \int h_i \mathbf{v}\nabla f_{i0} d\mathbf{v} + k_B \sum_i \int_{S_w} dS \int (\mathbf{v}\cdot\mathbf{n}) f_{i0} \frac{h_i^2}{2} d\mathbf{v} \qquad (21)$$

When writing the second integral in the right-hand side of equation (21) the integration is performed only over the lateral surface S_w of the capillary, since the integrals over face sections S in volume V in the stationary state are equal in magnitude and are opposite in sign.

The entropy production due to the interaction of radiation with the gas can be written as the sum of the entropy production due to the radiative decay of the excited level P_r and by induced transitions:

$$P_R = P_r + k_B \int dV \int \frac{\Gamma_m \chi^-(v_z)}{2}(f_{n0} - f_{m0})(h_n - h_m) d\mathbf{v} \qquad (22)$$

$$P_r = k_B \frac{\Gamma_m}{2} \int_V dV \int f_{m0}\left(h_m - h_n\right)^2 d\mathbf{v} \qquad (23)$$

Entropy production due to intermolecular collisions after linearization takes the form:

$$P_c = -k_B \sum_{i,j} \int dV \int f_{i0} h_i L_{ij}(h_i) d\mathbf{v} \qquad (24)$$

To find the meaning of the second term of the right side of expression (21) we calculate the entropy flux from wall to gas due to collisions of the gas particles with the surface of the capillary:

$$P_w = -k_B \sum_i \int_{S_w} dS \int (\mathbf{v}\cdot\mathbf{n}) f_i \ln f_i d\mathbf{v} = -k_B \sum_i \int_{S_w} dS \int (\mathbf{v}\cdot\mathbf{n}) f_{i0} \frac{h_i^2}{2} d\mathbf{v} + \int_{S_w} \frac{(\mathbf{q}\cdot\mathbf{n})}{T_w} dS \quad (25)$$

Here $(\mathbf{q}\cdot\mathbf{n}) = q_n^{gas}$ is the normal component of the heat flux density in gas on the wall of the capillary, T_w is the gas temperature at the surface of the capillary. The condition of continuity of the normal flux of heat in the steady state is performed at any point of the surface: $q_n^{gas} = q_n^{cap}$, where $q_n^{cap} = (\mathbf{q}^{cap}\cdot\mathbf{n})$ is the normal component of the heat flux density in the wall of the capillary.

The contribution of the second term in the right side of (25) in the entropy flux P_w depends not only on the thermal conductivity of the material of the capillary but also from the ratio of the radius of the capillary r_0 to its length L. In the limit $L \gg r_0$ its contribution becomes negligible under any coefficient of thermal conductivity of the wall. In this case in any cross section of the capillary in stationary conditions the state of local equilibrium is established when the normal component of vector of the heat flux density is zero and the temperature of the wall is equal to the temperature of the gas at any point of section. Therefore further, the second term in the right side of (25) we omit. Hence:

$$P_w = -k_B \sum_i \int_{S_w} dS \int (\mathbf{v}\cdot\mathbf{n}) f_{i0} \frac{h_i^2}{2} d\mathbf{v} \quad (26)$$

Substituting (21) - (24) and (26) into (20) we obtain the equation of entropy balance in the gas for the stationary state:

$$P = P_c + P_r + P_w = -k_B \sum_{i=m,n} \int_V dV \int h_i \mathbf{v}\nabla f_{i0} \, d\mathbf{v} + k_B \int dV \int \frac{\Gamma_m \chi^-(v_z)}{2} (f_{m0} - f_{n0})(h_n - h_m) d\mathbf{v}. \quad (27)$$

The entropy production inside the allocated volume of gas is caused by presence of the following sources of irreversible processes: intermolecular collisions (P_c), radiative decay of the excited level (P_r) and gas particles interaction with the capillary surface (P_w).

From property of symmetry of the collision integral [13] follows that $P_c \geq 0$. According to equation (23), we have $P_r \geq 0$. Using reciprocity for the scattering probability density (5) and Jensen's inequality for the continuous, strictly convex down functions [13], we get $P_w \geq 0$. Thus, the entropy production in the gas due to vector fluxes is nonnegative $P \geq 0$.

We transform the right-hand side of equation (27). We perform integration over the volume V like integration by capillary length L and cross-section area S. We take into account that in stationary conditions fluxes do not depend on the z coordinate, but the intensity of the radiation varies along the z axis due to absorption by gaseous medium. The dependence of z coordinate radiation intensity can be accounted for by introduction of the linear absorption coefficient β. Then the amplitude of the wave radiation electric field and, as a consequence, a square of the Rabi frequency proportional to the intensity of the radiation, in accordance with Bouguer law, can be written as

$$E(z) = E(0)\exp(-\frac{\beta}{2}z), \qquad G_{mn}^2(z) = G_{mn}^2(0)\exp(-\beta z). \quad (28)$$

where $G_{mn}(0)$ is the Rabi frequency associated with the intensity of radiation at the capillary entrance.

The integration in the right-hand side of eqn. (27) along the capillary within appropriate pressure and temperature limits with regard to (28), allows us to write this equation as:

$$P = \sum_k \mathbf{J}_k X_k \tag{29}$$

where the generalized fluxes $\mathbf{J}_k$ and forces X_k are defined as follows

$$\mathbf{J}_p = \sum_i \int dS \int v_z f_i d\mathbf{v} = \sum_i \int dS \int v_z f_{i0} h_i d\mathbf{v} \tag{30}$$

$$\mathbf{J}_T = \sum_i \int dS \int V_z \left(\frac{m_0 V^2}{2} + E_i \right) f_i d\mathbf{v} = k_B T_0 \sum_i \int dS \int v_z \left(c^2 - \frac{5}{2} + \varepsilon_i - \overline{\varepsilon} \right) f_{i0} h_i d\mathbf{v} \tag{31}$$

$$\mathbf{J}_S = k_B \sum_i \int dS \int \chi_S^- f_i \left(\frac{n_{j0}}{n_{i0}} - 1 \right) d\mathbf{v} = k_B \sum_i \int dS \int \chi_S^- f_{i0} h_i \left(\frac{n_{j0}}{n_{i0}} - 1 \right) d\mathbf{v} \tag{32}$$

$$X_P = -k_B \ln\left(\frac{p_2}{p_1} \right), \quad X_T = -\frac{1}{T_0} \ln\left(\frac{T_2}{T_1} \right), \quad X_S = \left(\frac{G_{mn}(0)}{k\overline{v}} \right)^2 \frac{1}{\beta} [1 - \exp(-\beta L)]. \tag{33}$$

$$\chi_S^- = \frac{\Gamma}{\left(\Gamma/k\overline{v} \right)^2 + \left(\Omega/k\overline{v} - c_z \right)^2} - \frac{\Gamma}{\left(\Gamma/k\overline{v} \right)^2 + \left(\Omega/k\overline{v} + c_z \right)^2},$$

$$\mathbf{V} = \mathbf{v} - \mathbf{U}, \quad k = \frac{2\pi}{\lambda}, \quad \overline{v} = \left(\frac{2k_B T_0}{m_0} \right)^{1/2}, \quad c_i = \frac{v_i}{\overline{v}}.$$

Here $\mathbf{V}$ is molecule own velocity with regard to the gas average macroscopic velocity $\mathbf{U}$, λ is the laser wavelength, S is the cross-section area of the capillary; p_1, T_1 and p_2, T_2 are pressure and temperature of the gas at the ends of the capillary.

The fluxes defined in eqn. (30) – (32) have an obvious physical meaning: $\mathbf{J}_p$ and $\mathbf{J}_T$ are respectively the mass and heat fluxes averaged over the cross-section of the capillary, $\mathbf{J}_S$ is the entropy flux calculated per unit length of the capillary.

Forces X_P and X_T for the case $(p_2 - p_1)/p_1 \ll 1$ and $(T_2 - T_1)/T_1 \ll 1$ were used in ref. [14] to describe non-isothermal gas motion in the channel. The force X_S is associated with the intensity of optical radiation, that is convenient because it is directly recorded in experiments [7].

4. Kinetic coefficients

Perturbation functions in linear approximation are a linear combination of generalized forces

$$h_i = h_i^p X_P + h_i^T X_T + h_i^S X_S \tag{34}$$

Substituting (34) into eqn. (30) – (32), we obtain to the fluxes

$$\mathbf{J}_l = \sum_k \Lambda_{lk} X_k \quad (k, l) = p, T, S \tag{35}$$

where the kinetic coefficients Λ_{lk} are defined by the following equations

$$\Lambda_{pp} = \left(\varphi_P, h_i^P \right), \qquad \Lambda_{pT} = \left(\varphi_p, h_i^T \right), \qquad \Lambda_{pS} = \left(\varphi_p, h_i^S \right),$$

$$\Lambda_{Tp} = k_B T_0 \left(\varphi_T, h_i^p \right), \qquad \Lambda_{TT} = k_B T_0 \left(\varphi_T, h_i^T \right), \qquad \Lambda_{TS} = k_B T_0 \left(\varphi_T, h_i^S \right), \tag{36}$$

$$\Lambda_{Sp} = k_B \left(\varphi_S, h_i^p \right), \qquad \Lambda_{ST} = k_B \left(\varphi_S, h_i^T \right), \qquad \Lambda_{SS} = k_B \left(\varphi_S, h_i^S \right).$$

$$\varphi_p = v_z, \qquad \varphi_T = v_z \left(c^2 - \frac{5}{2} + \varepsilon_i - \overline{\varepsilon} \right), \qquad \varphi_S = \chi_S^- (v_z) \left(\frac{n_{j0}}{n_{i0}} - 1 \right),$$

$$\left(\varphi_l, h_i \right) = \sum_{i=n,m} \int_S dS \int \varphi_l f_{i0} h_i d\mathbf{v}, \qquad l = p, T, S, \qquad i, j = n, m; \qquad i \neq j.$$

Here Λ_{lk} is the kinetic coefficient which determines contribution of generalized force X_k proportional component to the flux J_l. In so-called direct processes, force X_k defines the bound component of flux J_k, which is characterized by coefficient Λ_{kk}. The Poiseuille flux Λ_{pp} and the heat flux Λ_{TT} caused by the pressure and temperature gradients, respectively, are direct processes. The kinetic coefficient Λ_{SS} characterizes the entropy flux in the gas due to velocity selective radiation absorption by molecules. At the same time the forces X_k stimulate non-bound fluxes $J_i (i \neq k)$ characterized by kinetic cross-coefficients Λ_{ik}. In particular, temperature gradient stimulates gas motion along the capillary (thermal flux Λ_{pT}) and the entropy flux (thermo-entropic flux Λ_{ST}), and the pressure gradient causes heat flux (mechanocaloric effect Λ_{Tp}, [1]) and entropy flux (baro-entropic flux Λ_{Sp}). The action of resonant laser radiation leads to the drift of the gas Λ_{pS} and the heat flux Λ_{TS} which are directed across the baro- Λ_{Sp} and thermo-entropic Λ_{ST} fluxes respectively.

5. Onsager reciprocal relations
According to thermodynamics of nonequilibrium processes cross-coefficients in steady state must satisfy Onsager reciprocal relations:

$$\Lambda_{lk} = \Lambda_{kl} \tag{37}$$

To prove this we use the method proposed in ref. [12] and the modified in this study. Integrate the kinetic equation (11) for the odd perturbation functions h_i relatively to v_z along the length of the capillary. Substitute equation (34) for h_i in the obtained equations and then select summands under identical forces. As a result, we obtain the following system of equations for the functions h_i^l:

$$\mathbf{v}_\perp \frac{\partial h_i^P (\mathbf{v})}{\partial \mathbf{r}} - \sum_{j=n,m} L_{ij} \left(h_i^P (\mathbf{v}) \right) = \frac{\varphi_P (v_z)}{k_B L}$$

$$\mathbf{v}_\perp \frac{\partial h_i^T (\mathbf{v})}{\partial \mathbf{r}} - \sum_{j=n,m} L_{ij} \left(h_i^T (\mathbf{v}) \right) = \frac{T_0 \varphi_T (v_z)}{L} \tag{38}$$

$$\mathbf{v}_\perp \frac{\partial h_i^S (\mathbf{v})}{\partial \mathbf{r}} - \sum_{j=n,m} L_{ij} \left(h_i^S (\mathbf{v}) \right) - b_i h_m^S (\mathbf{v}) = \frac{\varphi_S (v_z)}{L}$$

In the equations (38) for h_i^P and h_i^T the terms describing the radiative decay of the excited level are absent. This is due to the fact that the pressure and temperature gradients anywhere in the gas do not stimulate local perturbations of equilibrium populations of the particles energy levels.

By analogy with eqn. (10) we present the perturbation functions h_i^l as the sum of the even h_i^{l+} and odd h_i^{l-} parts with respect to the transverse component of the molecules velocity vector $\mathbf{v}_\perp$. Then the equations (38) can be divided into even and odd with respect to the vector $\mathbf{v}_\perp$

$$\mathbf{v}_\perp \frac{\partial h_i^{P-}(v_z, \mathbf{v}_\perp)}{\partial \mathbf{r}} - \sum_{j=n,m} L_{ij}\left(h_i^{P+}(v_z, \mathbf{v}_\perp)\right) = \frac{\varphi_P(v_z)}{k_B L} \tag{39}$$

$$\mathbf{v}_\perp \frac{\partial h_i^{T-}(v_z, \mathbf{v}_\perp)}{\partial \mathbf{r}} - \sum_{j=n,m} L_{ij}\left(h_i^{T+}(v_z, \mathbf{v}_\perp)\right) = \frac{T_0 \varphi_T(v_z)}{L} \tag{40}$$

$$\mathbf{v}_\perp \frac{\partial h_i^{S-}(v_z, \mathbf{v}_\perp)}{\partial \mathbf{r}} - \sum_{j=n,m} L_{ij}\left(h_i^{S+}(v_z, \mathbf{v}_\perp)\right) - b_i h_m^{S+}(v_z, \mathbf{v}_\perp) = \frac{\varphi_S^-(v_z)}{L} \tag{41}$$

$$\mathbf{v}_\perp \frac{\partial h_i^{P,(T)+}(v_z, \mathbf{v}_\perp)}{\partial \mathbf{r}} = \sum_{j=n,m} L_{ij}\left(h_i^{P,(T)-}(v_z, \mathbf{v}_\perp)\right) \tag{42}$$

$$\mathbf{v}_\perp \frac{\partial h_i^{S+}(v_z, \mathbf{v}_\perp)}{\partial \mathbf{r}} - \sum_{j=n,m} L_{ij}\left(h_i^{S-}(v_z, \mathbf{v}_\perp)\right) - b_i h_m^{S-}(v_z, \mathbf{v}_\perp) = 0 \tag{43}$$

In (39) – (43) we take into account that the collision integrals for the odd (even) perturbation functions are the odd (even) functions of the velocity vector $\mathbf{v}_\perp$.

Reciprocity for thermal and mechanocaloric fluxes ($\Lambda_{pT} = \Lambda_{Tp}$) was proved in ref. [14]. Noteworthy that the proof of kinetic coefficients reciprocity that describe the transport processes is associated with exposure to radiation. Let us prove reciprocity relation for light-induced drift and baro-entropic flux $\Lambda_{pS} = \Lambda_{Sp}$. The equalities $\Lambda_{pT} = \Lambda_{Tp}$ and $\Lambda_{TS} = \Lambda_{ST}$ for cross-coefficients are proved similarly.

We multiply the equations (39) by h_i^S and the equations (41) by h_i^P. We subtract from the first obtained equation the second, summing over the states of the particles and integrating over velocity space and over the gas volume V. Taking into account the symmetry properties of the collision integral (commutativity of integral brackets) [13], we obtain

$$\Lambda_{pS} - \Lambda_{Sp} = k_B \sum_i \int_V dV \int f_{i0} \mathbf{v}_\perp \left(h_i^{S+} \nabla h_i^{P-} - h_i^{P+} \nabla h_i^{S-}\right) d\mathbf{v} + k_B \sum_i \int dV \int f_{i0} h_i^{P+} b_i h_m^{S+} d\mathbf{v} \tag{44}$$

The first term on the right side of equation (44) taking (42) and (43) is equal to

$$k_B \sum_i \int_V dV \int f_{i0} \mathbf{v}_\perp \left(h_i^{S+} \nabla h_i^{P-} - h_i^{P+} \nabla h_i^{S-}\right) d\mathbf{v} =$$
$$= k_B \sum_{i,j=m,n} \int dV \int f_{i0} \left[\mathbf{v}_\perp \nabla (h_i^{P-} h_i^{S+} - h_i^{P+} h_i^{S-}) - h_i^{P-} b_i h_m^{S-}\right] d\mathbf{v} \tag{45}$$

We substitute the equation (45) into eqn. (44)

$$\Lambda_{pS} - \Lambda_{Sp} = k_B \sum_i \int_V dV \int f_{i0} \mathbf{v}_\perp \nabla \left(h_i^{S+} h_i^{P-} - h_i^{P+} h_i^{S-} \right) d\mathbf{v} +$$
$$+ k_B \sum_i \int dV \int f_{i0} b_i (h_i^{P+} h_m^{S+} - h_i^{P-} h_m^{S-}) d\mathbf{v} \tag{46}$$

According to the theorem of the equivalence of the infinitesimal functions [15], the second term on the right-hand side of (46) is the infinitesimal magnitude of the higher (fourth) order of smallness and should be omitted. After applying Gauss transform to the first term in right part of eqn. (46), we get

$$\Lambda_{pS} - \Lambda_{Sp} = k_B \sum_i \int_{S_w} dS \int f_{i0} (\mathbf{v} \cdot \mathbf{n})(h_i^{S-} h_i^{P+} - h_i^{P-} h_i^{S+}) d\mathbf{v} \tag{47}$$

Demonstrate that the integral in the right-hand side of (47) vanishes.

On the surface of the capillary the even h_i^{l+} and odd h_i^{l-} parts of perturbation functions have discontinuity and satisfy the following conditions:

$$h_i^{l+}(\mathbf{v}) = \frac{h_i^l(-\mathbf{u}) + h_{iw}^l(\mathbf{u})}{2} \tag{48}$$

$$h_i^{l-}(\mathbf{v}) = \mp \frac{h_i^l(-\mathbf{u}) - h_{iw}^l(\mathbf{u})}{2} = -\frac{|\mathbf{v} \cdot \mathbf{n}|}{(\mathbf{v} \cdot \mathbf{n})} \frac{h_i^l(-\mathbf{u}) - h_{iw}^l(\mathbf{u})}{2} \tag{49}$$

$$\mathbf{u} = \mathbf{v} \, \text{sgn}(\mathbf{v} \cdot \mathbf{n}), \qquad l = P, T, S \, .$$

Here $h_{iw}^l(\mathbf{u})$ are perturbation functions for the molecules reflected from the surface of capillary. The minus sign after the first equality of eqn. (49) is used at $(\mathbf{v} \cdot \mathbf{n}) > 0$, plus sign at $(\mathbf{v} \cdot \mathbf{n}) < 0$.

Eliminating from (48) and (49) $h_i^l(-\mathbf{u})$, we obtain the connection between the functions $h_i^{l-}(\mathbf{v})$ and $h_i^{l+}(\mathbf{v})$ at the boundary surface:

$$(\mathbf{v} \cdot \mathbf{n}) h_i^{l-}(\mathbf{v}) = -|\mathbf{v} \cdot \mathbf{n}|(h_i^{l+}(\mathbf{v}) - h_{iw}^l(\mathbf{u})) \tag{50}$$

Substituting (48) and (50) into (47) for the case $l = P, S$ we obtain the equality

$$\Lambda_{pS} - \Lambda_{Sp} = \frac{k_B}{2} \sum_i \int_{S_w} dS \int f_{i0} |\mathbf{v} \cdot \mathbf{n}| \left[h_i^P(-\mathbf{u}) h_{iw}^S(\mathbf{u}) - h_i^S(-\mathbf{u}) h_{iw}^P(\mathbf{u}) \right] d\mathbf{v} \tag{51}$$

Consider the integral

$$Z = \frac{k_B}{2} \sum_i \int_{S_w} dS \int h_i^P(-\mathbf{u}) f_{i0} |\mathbf{v} \cdot \mathbf{n}| h_{iw}^S(\mathbf{u}) d\mathbf{v} \tag{52}$$

Replace $h_{iw}^S(\mathbf{u})$ with (9) and integrate over the half space of the velocities $\mathbf{v} \cdot \mathbf{n} > 0$:

$$Z = \frac{k_B}{2} \sum_{i,j} \int_{S_w} dS \int_{\mathbf{v} \cdot \mathbf{n} > 0} h_i^P(-\mathbf{v}) d\mathbf{v} \int_{\mathbf{v}' \cdot \mathbf{n} < 0} |\mathbf{v}' \cdot \mathbf{n}| f_{j0}(\mathbf{v}') h_j^S(\mathbf{v}') \cdot B(j, \mathbf{v}' \to i, \mathbf{v}) d\mathbf{v}' \tag{53}$$

Reciprocity for the scattering kernel (5) gives the equation (53) the form:

$$Z = \frac{k_B}{2} \sum_{i,j} \int_{S_w} dS \int_{\mathbf{v}'\cdot\mathbf{n}<0} h_j^S(\mathbf{v}')d\mathbf{v}' \int_{\mathbf{v}\cdot\mathbf{n}>0} |\mathbf{v}\cdot\mathbf{n}| f_{i0}(\mathbf{v}) h_i^P(-\mathbf{v}) B(i,-\mathbf{v} \to j,-\mathbf{v}')d\mathbf{v} \tag{54}$$

We replace $\mathbf{v}' \to -\mathbf{v}$, $-\mathbf{v}' \to \mathbf{v}$, $i \to j$, $j \to i$. Then

$$Z = \frac{k_B}{2} \sum_{i,j} \int_{S_w} dS \int_{\mathbf{v}\cdot\mathbf{n}>0} h_i^S(-\mathbf{v})d\mathbf{v} \int_{\mathbf{v}'\cdot\mathbf{n}<0} |\mathbf{v}'\cdot\mathbf{n}| f_{j0}(\mathbf{v}') h_j^P(\mathbf{v}') \cdot B(j,\mathbf{v}' \to i,\mathbf{v})d\mathbf{v}' \tag{55}$$

When returning to complete space of velocities, with regard to (9) we get:

$$Z = \frac{k_B}{2} \sum_{i,j} \int_{S_w} dS \int h_i^S(-\mathbf{u})d\mathbf{v} \int_{\mathbf{v}'\cdot\mathbf{n}<0} |\mathbf{v}'\cdot\mathbf{n}| f_{j0}(\mathbf{v}') h_j^P(\mathbf{v}') \cdot B(j,\mathbf{v}' \to i,\mathbf{v})d\mathbf{v}' =$$

$$= \frac{k_B}{2} \sum_i \int_{S_w} dS \int f_{i0} |\mathbf{v}\cdot\mathbf{n}| h_i^S(-\mathbf{u}) h_{iw}^P(\mathbf{u})d\mathbf{v} \tag{56}$$

Substituting (56) into (51) we get $\Lambda_{pS} = \Lambda_{Sp}$, Q.E.D.

We note that the reciprocity relations are proved for arbitrary Knudsen (*Kn*) numbers and for any laws of interaction of the molecules with each other and with boundary surface.

6. Non-equilibrium stationary states

Discuss now the nonequilibrium stationary processes when the boundary conditions imposed on the system do not allow it to reach the equilibrium state. These processes have been considered in the linear approximation in ref. [16]. In stationary states the entropy production unlike the equilibrium states does not disappear. According to Prigogine's principle [1] in the stationary weakly non-equilibrium states the entropy production is minimal. If *n* of thermodynamic forces act on the system with fixed *k* forces then steady state of *k*-order is established in the system. Now the fluxes associated with other *n-k* forces are equal to zero.

Consider the possible stationary states. We start with the well-known effects.

6.1. Thermomolecular pressure difference (*TPD-effect*) [1, 14, 17].
Take the gas is in a closed system, which consists of two volumes connected by a capillary. If the radiation is absent, $X_S = 0$, and the temperature difference at the ends of the capillary is fixed, $X_T = const$, then the pressure drop is established at the ends of the capillary. This phenomenon is known as thermomolecular pressure difference. In this case, the gas flow in the capillary is equal to zero, $J_p = 0$, i.e. the thermal transport is compensated by the Poiseuille flux. With the use of (33), (35) and (36) we get [17]:

$$\frac{p_2}{p_1} = \left(\frac{T_2}{T_1}\right)^{\gamma_T}, \qquad \gamma_T = -\frac{1}{k_B T_0}\frac{\Lambda_{pT}}{\Lambda_{pp}} \tag{57}$$

where γ_T is the TPD index, defined as the ratio of the thermal creep of the Poiseuille flux. The value γ_T depends on the parameters of gas interaction with the surface of the capillary and *Kn* number. It is known [17] that in a slip-flow regime $\gamma_T \sim Kn^2$, and in free-molecular limit $(Kn \gg 1)$ in the case of specular-diffuse reflection of molecules from the capillary surface $\gamma_T = 1/2$.

6.2. Mechanocaloric effect [1, 14].

This effect is responsible for the thermal energy transfer under isothermal movement of the gas. If the radiation is absent, $X_S = 0$ and the gas temperature is uniform, $X_T = 0$, then, according to (35), the heat flux is defined as

$$J_T = -\Lambda_{Tp} k_B \ln\left(\frac{p_2}{p_1}\right).$$ (58)

The magnitude of q_p^* indicates the amount of heat that is transferred per unit time through the cross-section of the capillary under the action of pressure drop calculated per molecule:

$$q_p^* = \frac{J_T}{J_p} = \frac{\Lambda_{Tp}}{\Lambda_{pp}}.$$ (59)

Given the symmetry of cross kinetic coefficients, $\Lambda_{Tp} = \Lambda_{pT}$ from (57) and (59) we find connection between values $q_p^\bullet$ and γ_T

$$q_p^* = -k_B T_0 \gamma_T.$$ (60)

6.3. Light-induced pressure difference (LIPD-effect) [7, 18] is the emergence of the gas pressure drop at the ends of closed capillary under the action of resonant optical radiation.

Gas with uniform response to temperature ($X_T = 0$) is located in a closed capillary. The ends of the capillary can freely pass the laser radiation. The light-induced drift of the gas will stimulate the pressure redistribution along the capillary. The pressure inhomogeneity will cause the Poiseuille flux. In the steady state the light-induced drift will be compensated by the Poiseuille flux:

$$J_p = \Lambda_{pp} X_p + \Lambda_{pS} X_S = 0.$$ (61)

From here taking into account (33) we obtain

$$\frac{p_2}{p_1} = \exp(\gamma_{ST}), \qquad \gamma_{ST} = \frac{1}{\beta k_B}\left(\frac{G_{mn}(0)}{k\overline{\upsilon}}\right)^2 \left(\frac{\Lambda_{pS}}{\Lambda_{pp}}\right)\left[1 - \exp(-\beta L)\right].$$ (62)

In the case of an optically thin medium ($\beta L \ll 1$) expression for the parameter γ_{ST} simplifies and becomes

$$\gamma_{ST} = \frac{L}{k_B}\left(\frac{\Lambda_{pS}}{\Lambda_{pp}}\right)\left(\frac{G_{mn}(0)}{k\overline{\upsilon}}\right)^2.$$ (63)

The value of the index γ_{ST} depends on the parameters of the gas interaction with the capillary surface, detuning frequency radiation from the center of the absorption line, the intensity and wavelength of the radiation, as well as on *Kn* number. As shown in [19], in a slip-flow regime $\gamma_{ST} \sim Kn^2$, and in the free-molecular regime γ_{ST} does not depend on *Kn*.

6.4. Thermo-optical pressure difference (TOPD-effect) [19] is the phenomenon of the establishment of the gas pressure drop at the ends of closed capillary under the joint action of the resonant optical radiation and temperature inhomogeneity. This is a stationary state of second order in which two forces: X_T and X_S are fixed.

Longitudinal pressure gradient arises in such a system as a result of the light-induced drift of gas and thermal creep. In turn it will cause a Poiseuille flux. As a result of competition of non-equilibrium processes in the system is settles in some steady state of dynamic equilibrium. It will be the state with a minimum entropy production at which the gas flux $\mathbf{J}_p$ associated with the unfixed force X_p must be equal to zero:

$$\mathbf{J}_p = \Lambda_{pp} X_p + \Lambda_{pT} X_T + \Lambda_{pS} X_S = 0 \quad . \tag{64}$$

From here follows:

$$\ln\left(\frac{p_2}{p_1}\right) = -\frac{1}{k_B T_0}\left(\frac{\Lambda_{pT}}{\Lambda_{pp}}\right)\ln\left(\frac{T_2}{T_1}\right) + \frac{1}{k_B}\left(\frac{\Lambda_{pS}}{\Lambda_{pp}}\right) X_S \quad . \tag{65}$$

Taking into account (33) we obtain the following expression for the pressure drop of gas at the ends of the capillary

$$\frac{p_2}{p_1} = \left(\frac{T_2}{T_1}\right)^{\gamma_T} \exp\left(\gamma_{ST}\right) \quad . \tag{66}$$

6.5. Optocaloric effect is the light–induced heat transfer.

Pressure and temperature of the gas are uniform, $X_p = 0$, $X_T = 0$, the radiation force is fixed $X_S = const$. Then according to (35) the heat flux and the drift gas are governed only by the radiation force X_S. Magnitude of q_S^* characterizes the quantity of heat which is transferred per unit time through a cross-section of the capillary under action the resonant optical radiation in calculation per molecule:

$$q_S^* = \frac{\mathbf{J}_T}{\mathbf{J}_p} = \frac{\Lambda_{TS}}{\Lambda_{PS}} \quad . \tag{67}$$

6.6. Light-induced baro- and thermo-effect [9] is phenomenon of simultaneous occurrence of pressure and the gas temperature difference at the ends of a closed thermally insulated capillary under exposure to resonant optical radiation. This is a stationary state of the first order when only the force of X_S is fixed. Light-induced gas drift and light-induced heat flux cause longitudinal pressure and temperature gradients in the system. These gradients, in turn, cause the own fluxes that will compete with light-induced fluxes. In the system some steady state of dynamic equilibrium is eventually established. According to Prigogine's principle [1], it will be state with a minimum entropy production at which the fluxes $\mathbf{J}_P$ and $\mathbf{J}_T$, associated with unfixed forces X_p and X_T must be equal to zero:

$$\mathbf{J}_p = \Lambda_{pp} X_p + \Lambda_{pT} X_T + \Lambda_{pS} X_S = 0, \tag{68}$$

$$\mathbf{J}_T = \Lambda_{Tp} X_p + \Lambda_{TT} X_T + \Lambda_{TS} X_S = 0.$$

Equations for the light-induced differential pressure and temperature follow from here:

$$-k_B \ln\left(\frac{p_2}{p_1}\right) = X_S \left(\Lambda_{pT}\Lambda_{TS} - \Lambda_{pS}\Lambda_{TT}\right)/\Delta_0$$

$$-\frac{1}{T_0}\ln\left(\frac{T_2}{T_1}\right) = X_S\,(\Lambda_{Tp}\Lambda_{pS} - \Lambda_{pp}\Lambda_{TS})\big/\Delta_0 \tag{69}$$

$$\Delta_0 = \Lambda_{pp}\Lambda_{TT} - \Lambda_{Tp}\Lambda_{pT}$$

We see that longitudinal redistribution of gas pressure in the capillary is caused by light-induced drift (LID) and thermal transport, and also depends on heat fluxes. Thermal fluxes lead to the inhomogeneity of temperature that, in turn, is the cause of the thermal transport. The temperature difference is due to the light-induced heat transfer and the mechanocaloric flux, and depends also on Poiseuille flux and light-induced drift. Fluxes of the gas make the pressure gradient and hence the mechanocaloric flux.

Thus the resonant laser radiation can perform not only function of the piston compressing the gas, but also a sort of heat engine, capable to redistribute thermal energy in a gas.

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