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**KINETIC THEORY OF
RADIOMETRIC PHOTOPHORESIS
IN A KNUDSEN GAS**

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KINETIC THEORY OF RADIOMETRIC PHOTOPHORESIS
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by

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Abstract

The kinetic theory of a small particle with arbitrary temperature distribution on its surface suspended in a Knudsen gas enclosed by an unequally heated boundaries has been established according to the revised law of thermal transpiration developed recently by Wu. The external force acting on the particle is mainly due to the radiometric force caused by the incident and the reflecting molecules in the absence of thermal creep effect in this limiting regime. Several particle shapes have been considered, such as a small disk, flat plate, sphere and cube presented in a cylinder with uniform temperature gradient in a thermal transpiration system.

I. INTRODUCTION

In 1825 Fresnel first discovered that a small body suspended in a gas is sometimes set into motion when light falls upon it. Crookes (1874-1878) studied this effect by his well known radiometer in which the body takes the form of vanes blackened on one side, carried by a crossbar which rotates when illuminated by radiation. Ehrenhaft (1914) observed an analogous motion of microscopic particles so called photophoresis suspended in a gas and found that some particles move toward the light, others against it.⁽¹⁾

The laws governing all these phenomena appear to be quite similar. It has been well understood that in all cases the light acts by heating the suspended body. The same effects can be produced by establishing in the gas such a temperature gradient as will give rise to the same temperature differences at the surface of the suspended body. The general rule is that hot surfaces behave as if repelled by the gas. The movements toward the light which are often observed in photophoresis are ascribed to greater heating of the far side of a transparent particle; this latter effect has been successfully imitated with a radiometer carrying disk of molybdenite, one of whose surfaces was fresher than the other.

Various opinions have been expressed as to the origin of the radiometric force. As Maxwell⁽²⁾ suggested in 1879 that most radiometric phenomena is due to the thermal creep of the gas over an unequally heated solid surface. Several theoretical and experimental studies have confirmed this explanation.

The existence of such thermal creep flow in the gas was shown by Gerlach and Schütz in 1932.⁽³⁾ They suspended a tiny vanelet near a radiometer

and observed that it was deflected by the action of the streaming gas. Similar experiment in 1924, Czerny and Hettner⁽⁴⁾ reached the same conclusion.

Very recently, Wu studied the radiometer in the Knudsen regime and found the thermal creep flow in a closed system does not exist at all in this limiting regime.^(5,6) However, the radiometric force in Knudsen gas exists only due to the incident and the reflected molecules in this stationary state system. In this paper we shall study the external force on a particle suspended in a closed system in a Knudsen gas regime. A radiometric drag on the particle exists in the absence of viscous force due to the incident molecular force from the unequally heated boundary and the reflected molecular force from nonuniform distribution of temperature on the particle.

II. THEORY OF RADIOMETRIC FORCE

Qualitatively the thermal creep theory of radiometric action is comparatively successful; however, a quantitative calculation of the viscous force presents a rather difficult problem. The viscous force theories of Epstein⁽⁷⁾ and Sexe⁽⁸⁾ were developed based on the assumption of thermal creep flow. In general these theories are certainly not valid in the Knudsen gas. In this paper, we shall establish a kinetic theory of this phenomenon in the Knudsen gas regime according to the revised law of thermal transpiration established recently by Wu.⁽⁹⁾ The flow field has been proved to be stationary everywhere in a closed system, and the distribution function in the flow field is known exactly, provided that the geometry of the boundary is given. We shall consider a small particle (or a photophoresis) with fully or partially accommodated coefficients on its surface. The temperature distribution on the surface is given. This particle is suspended in a stationary system in which the density of the gas is so low that the mean free path of the gas is much greater than the dimension of the local surrounding boundaries.

We first consider a Knudsen gas in a flow field enclosed by an arbitrary boundary with a given temperature distribution and accommodation coefficient on its surfaces. The condition of equilibrium according to the revised theory of thermal transpiration states that PI/\sqrt{T} (or $n\sqrt{TI}$) is an invariant throughout the system (Figure 1). The new function I (isotropy) is a measure of the isotropic characteristics of the velocity distribution function of the gas in the flow field. If a small particle is present at a given position in the system and the temperature distribution on the body surface is known, and the thermal accommodation coefficient on the body surface and

the boundary of the system is assumed to be unity, then, the distribution function of the gas in the flow field can be described in the following form:

$$F_{\varphi\theta} = n_{\varphi\theta} f_{\varphi\theta} = K \left(\frac{1}{2\pi R} \right)^{3/2} \left(\frac{1}{T_{\varphi\theta}} \right)^2 e^{-\frac{c^2}{2RT_{\varphi\theta}}} \quad (2.1)$$

In an axisymmetric boundary condition, this distribution function reduces to the form,

$$F_{\varphi} = n_{\varphi} f_{\varphi} = K \left(\frac{1}{2\pi R} \right)^{3/2} \left(\frac{1}{T_{\varphi}} \right)^2 e^{-\frac{c^2}{2RT_{\varphi}}} \quad (2.2)$$

where,

$$K = n_{\varphi\theta} \sqrt{T_{\varphi\theta}} = n_{\varphi} \sqrt{T_{\varphi}} I = \frac{PI}{\sqrt{T}} \frac{1}{mR} = \frac{N}{4\pi \int_{V_{\varphi\theta}} \frac{1}{\sqrt{T_{\varphi\theta}}} \sin\varphi d\varphi d\theta d\vec{x}} \quad (2.3)$$

or

$$K = \frac{E}{mC_v \frac{1}{4\pi} \int_{V_{\varphi\theta}} \sqrt{T_{\varphi\theta}} \sin\varphi d\varphi d\theta d\vec{x}} \quad (2.4)$$

in which N and E are the total number of molecules and total kinetic energy in the system respectively.

The stress tensor of the gas will be,

$$P_{ij} = m \int_{\varphi\theta c} V_i V_j n_{\varphi\theta} f_{\varphi\theta} d\vec{\omega} = P\delta_{ij} - \tau_{ij} \quad (2.5)$$

and the momentum transfer in the direction of X axis on a surface with the normal vector \vec{n} is

$$P_{nx} = -m \iiint_{c\varphi\theta} V_n V_x n_{\varphi\theta} f_{\varphi\theta} d\vec{\omega} \quad (2.6)$$

and the normal stress on a surface is,

$$P_{nn} = m \iiint_{c\varphi\theta} V_n^2 n_{\varphi\theta} f_{\varphi\theta} d\vec{\omega} \quad (2.7)$$

However, for a particle, the distribution function of the gas at the surface is described separately in the two half space range; i.e.,

$$F_{(+)} = K \left(\frac{1}{2\pi R} \right)^{3/2} \left(\frac{1}{T_s} \right)^2 e^{-\frac{c^2}{2RT_s}} \quad (2.8)$$

$$F_{(-)} = n_{\varphi\theta} f_{\varphi\theta} = K \left(\frac{1}{2\pi R} \right)^{3/2} \left(\frac{1}{T_{\varphi\theta}} \right)^2 e^{-\frac{c^2}{2RT_{\varphi\theta}}} \quad (2.9)$$

where the (+) denotes the reflected molecules from the surface and the (-) denotes the incident molecules to the surface; and T_s is the temperature on the body surface.

III. RADIOMETRIC PHOTOPHORESIS

Let us consider a particle suspended in a Knudsen gas on the axis of a long cylinder which is either closed at the ends or open to a large closed container in a stationary state system (Figure 2). The mean free path of the gas is much greater than the diameter of the cylinder. Thus the invariant K exists throughout the cylinder as well as the container. The temperature distribution along the cylinder surface is assumed to be linear and the temperature difference between two ends is comparatively smaller than that of the lower temperature side. Thus the directional temperature (T_φ) on the axis of the cylinder is axisymmetric. Therefore,

$$T_{\varphi\theta} = T_\varphi(x) = T_x + \frac{\Delta T}{\Delta x} x' \text{ and } x' = \gamma \cot \varphi. \quad (3.1)$$

Let $\frac{\Delta T}{\Delta x} = G$, we have

$$F_{(-)}(x) = n_{\varphi\theta} f_{\varphi\theta(-)} = K \left(\frac{1}{2\pi R} \right)^{3/2} \left(\frac{1}{T} \right)_\varphi^2 e^{-\frac{c^2}{2RT_\varphi}} \quad (3.2)$$

where

$$T_\varphi = T_x + G\gamma \cot \varphi. \quad (3.3)$$

According to the velocity distribution function, it is evident that the Knudsen gas is stationary in the flow field. For this reason, there is no viscous force or thermal creep force existing on the particle in a Knudsen gas. The only force acting on the particle is the radiometric force or the momentum transfer due to the incident and reflecting molecules. It is interesting to see that the radiometric force due to temperature gradient of adjacent plates will

cause a force which is in the opposite direction of a viscous force in transition flow. In other words, the particle will move in the opposite direction while the density of the gas will approach to the free molecular regime. We shall study two kinds of particles: (a) those on which the temperature distribution is uniform (highly conductive) and (b) those on which the temperature distribution is non-uniform. In both cases the particles are assumed to be contained in a long cylinder with constant temperature gradient on its boundary.

Let us first consider several different particles with uniform temperature on the surfaces as follows (Figure 3):

(1) a thin disk or plate

(a) parallel to the axis

The radiometric force acting along the x-axis will be caused only by the incident molecules on this plate. Due to the symmetric condition the shear force on both side per unit area will be

$$P_{zx}(-) = 2m \overline{nWU_{\vec{w} \geq 0}} = -2m \int_{c=0}^{\infty} \int_{\varphi=0}^{\pi} \int_{\theta=0}^{\pi} WU n_{\varphi\theta} f_{\varphi\theta} d\vec{w} \quad (3.4)$$

Since

$$\vec{V} = (U, V, W) = (C \cos \varphi, C \sin \varphi \cos \theta, C \sin \varphi \sin \theta) d\vec{w} = c^2 \sin \varphi d\varphi d\theta dc,$$

$$n_{\varphi\theta} f_{\varphi\theta} = n_{\varphi} f_{\varphi} \quad \text{and} \quad T_{\varphi\theta} = T_{\varphi} = T_X + GY \cot \varphi,$$

we have,

$$\begin{aligned} P_{zx}(-) &= -2m \int_{c=0}^{\infty} \int_{\varphi=0}^{\pi} \int_{\theta=0}^{\pi} K \left(\frac{1}{2\pi R} \right)^{3/2} \frac{1}{T_{\varphi}} e^{-\frac{c^2}{2RT_{\varphi}}} c^4 \cos \varphi \sin^2 \varphi \sin \theta d\varphi d\theta dc \\ &= -\frac{3}{\pi} mKR \int_0^{\pi} \sqrt{T_{\varphi}} \cos \varphi \sin^2 \varphi d\varphi \end{aligned} \quad (3.5)$$

By the assumption that $G \gamma \cot \varphi < T_x$ for $\varphi > \epsilon$ and $\pi - \varphi > \epsilon$

Equation (3.5) becomes

$$P_{zx}(-) = - \frac{3}{\pi} mKR \int_0^\pi \left(\sqrt{T_x} + \frac{1}{2} \frac{G \gamma \cot \varphi}{\sqrt{T_x}} \right) \cos \varphi \sin^2 \varphi d\varphi + \text{h.o.t.} \quad (3.6)$$

Since the first term in the integral vanishes and the higher order terms are negligible, we obtain

$$P_{zx}(-) = - \frac{mKR\gamma}{\pi \sqrt{T_x}} G \quad (3.7)$$

Notice that $P_{zx}(-) \propto -G$, which shows that the radiometric shear force is proportional to the negative temperature gradient. On the contrary, the viscous force caused by the thermal creep effect in the transition flow is directly proportional to the temperature gradient. The radiometric drag can be expressed in the form

$$D_x(-) = - \frac{mKR\gamma}{\pi \sqrt{T_x}} GA, \quad (3.8)$$

where A is the area of the disk or plate.

If the temperature gradient is not large so that the isotropy in the gas is very close to unity, then, $K = n\sqrt{T} I \approx n \sqrt{T_x}$.

Therefore, the drag becomes,

$$D_x(-) = - \frac{mnR\gamma}{\pi} GA = - \frac{mnR}{\pi} \frac{\Delta T}{\Delta(x/\gamma)} A \quad (3.9)$$

The drag force is independent of the local temperature but depends only on the local density and the temperature gradient.

(b) normal to the axis

The radiometric force caused by the incident molecules is normal to the surface from both sides, i.e.,

$$\begin{aligned}
 P_{xx}(-) &= - \overline{mnU_{u \geq 0}^2} + \overline{mnU_{u \leq 0}^2} \\
 &= -m \int_0^\infty \int_0^{\pi/2} \int_0^{2\pi} n_{\varphi} f_{\varphi} c^4 \cos^2 \varphi \sin \varphi d\varphi d\theta dc \\
 &\quad + m \int_0^\infty \int_{\pi/2}^\pi \int_0^{2\pi} n_{\varphi} f_{\varphi} c^4 \cos^2 \varphi \sin \varphi d\varphi d\theta dc \\
 &= - \frac{mKRG\gamma}{\sqrt{T_x}} A + \text{h.o.t.} \tag{3.10}
 \end{aligned}$$

By neglecting the higher order terms, we have the drag force in the x direction,

$$\begin{aligned}
 D_x(-) &= - \frac{mKRG\gamma}{\sqrt{T_x}} A \approx mnR\gamma GA \\
 &= - mnR \frac{\Delta T}{\Delta(x/\gamma)} A \tag{3.11}
 \end{aligned}$$

The drag force in this case is π times higher than that of the previous example.

(2) a small sphere

If the sphere is very small compared with the dimension of the system, then the total momentum incident on the particle can be calculated as a point sink on the body. The total resultant force per unit normal area (i.e. $A_n = \frac{\pi}{4} d^2$)

is in the X direction, i.e.,

$$\begin{aligned}
 P_{nx}(-) &= - m \overline{ncU} \\
 &= - m \int_{c=0}^{\infty} \int_{\varphi=0}^{\pi} \int_{\theta=0}^{2\pi} n_{\varphi} f_{\varphi} c^4 \cos\varphi \sin\varphi d\varphi d\theta dc \\
 &= - \frac{3\pi}{8} \frac{mKRG\gamma}{\sqrt{T_x}} + \text{h.o.t.}
 \end{aligned} \tag{3.12}$$

By neglecting the higher order term, we obtain the radiometric drag in the X direction as follows,

$$\begin{aligned}
 D_x(-) &= P_{nx}(-) \times \frac{\pi}{4} d^2 = - \frac{3\pi}{8} \frac{mKRG\gamma}{\sqrt{T_x}} \left(\frac{\pi}{4} d^2 \right) \\
 &\approx \frac{3\pi}{8} mnR \frac{\Delta T}{\Delta(x/\gamma)} \times \frac{\pi d^2}{4}
 \end{aligned} \tag{3.13}$$

(3) a small cube

Similarly, the radiometric drag force on a small cube can be obtained by superposing those of two parallel plates and a normal plate, i.e.,

$$D_x(-) \approx - \left(\frac{2}{\pi} + 1 \right) mnR \frac{\Delta T}{\Delta(x/\gamma)} A \tag{3.14}$$

where A is the surface area of one side of the cube.

Let us discuss the effect of non-uniform temperature distribution on the particles. If the temperature distribution is not uniform on the particles, but homogeneous in a certain part of the surface, the density distribution of the gas away from the surface in this part will also be homogeneous according to the

revised theory of thermal transpiration. Therefore, the external force acting on this surface is always normal to the surface and equal to one-half of the pressure in an equilibrium condition with corresponding temperature and density reflected from the surface. Consequently, it is quite easy to calculate the radiometric force due to reflecting molecules.

It is evident that the drag forces in the X direction, as shown in Figure 3, are as follows,

(i) for a thin plate normal to the axis

$$\begin{aligned} D_{x(+)} &= \left(\frac{mRn_1 T_1}{2} - \frac{mRn_2 T_2}{2} \right) A \\ &= \frac{mRK}{2} (\sqrt{T_1} - \sqrt{T_2}) A \cong \frac{mRn\sqrt{T_x}}{2} (\sqrt{T_1} - \sqrt{T_2}) A \end{aligned} \quad (3.15)$$

(ii) for a small sphere,

$$D_{x(+)} = \frac{mRK}{2} (\sqrt{T_1} - \sqrt{T_2}) \left(\frac{\pi d^2}{4} \right) \cong \frac{mRn\sqrt{T_x}}{2} (\sqrt{T_1} - \sqrt{T_2}) \left(\frac{\pi d^2}{4} \right) \quad (3.16)$$

Certainly, the resulting forces on the particles should be the sum of the present solution and the previous one, where both effects are in the same order of magnitude. If the accommodation coefficient on the surface is not unity, the temperatures in equations (3.15) and (3.16) should be replaced by T_1' and T_2' , which are the temperatures of the reflected molecules at surfaces 1 and 2 respectively, according to the definition of the diffuse reflection law.

IV. DISCUSSION

It has been shown that in the absence of thermal creep flow, the particle with uniform temperature in a thermal transpiration system will move from higher temperature region to lower temperature region. The radiometric force on the body is in the opposite direction to the thermal creep force. However the temperature difference at the surface of the suspended body will create a movement toward the lower temperature side. In certain cases the thermal creep flow may exist in a locally free-molecular region. In such cases one has to find the exact distribution function in the flow field in order to solve the complete problem.

In view of the many varieties of the problems with different boundary conditions, the present theory with its unique treatment is regarded as very encouraging. However, in the near free molecular regime or higher order transition regimes, the present theory could be served as a first order approximation in an iteration process.

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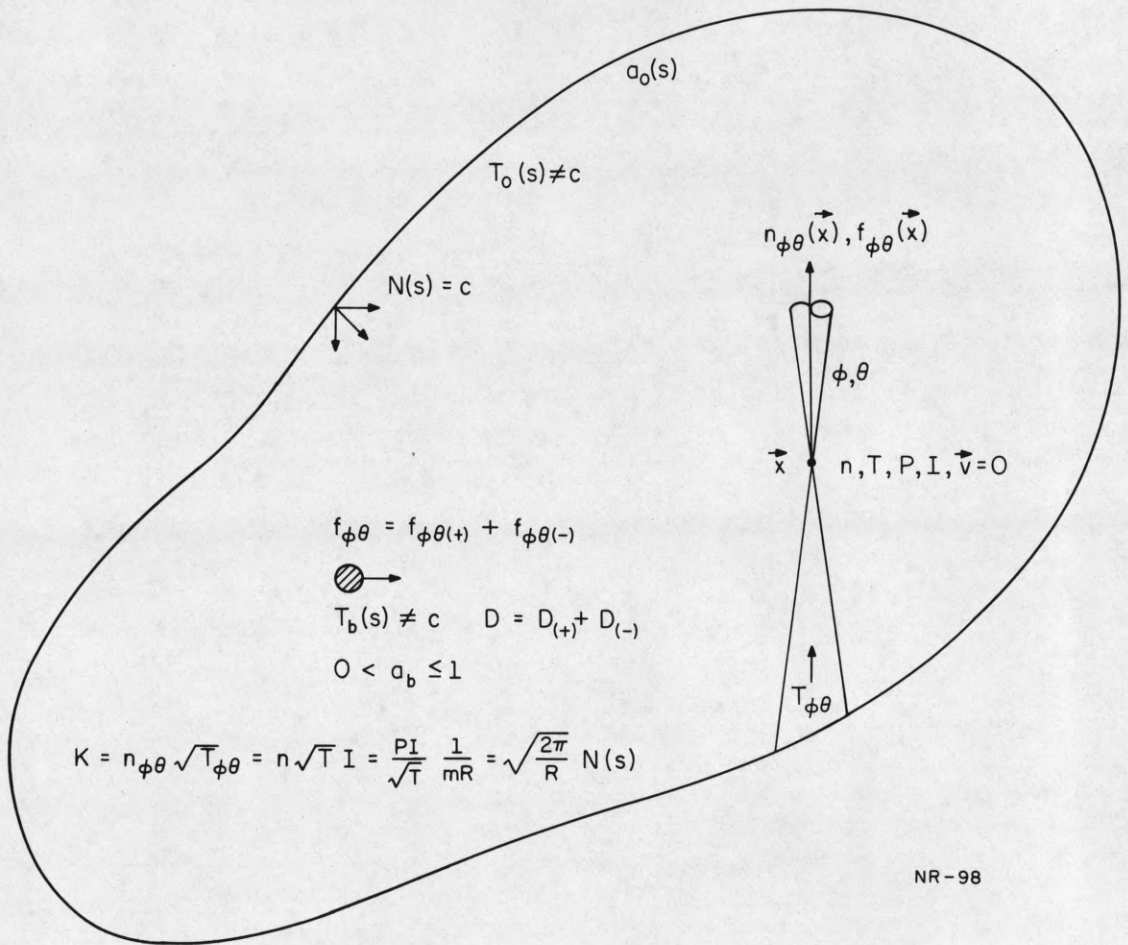
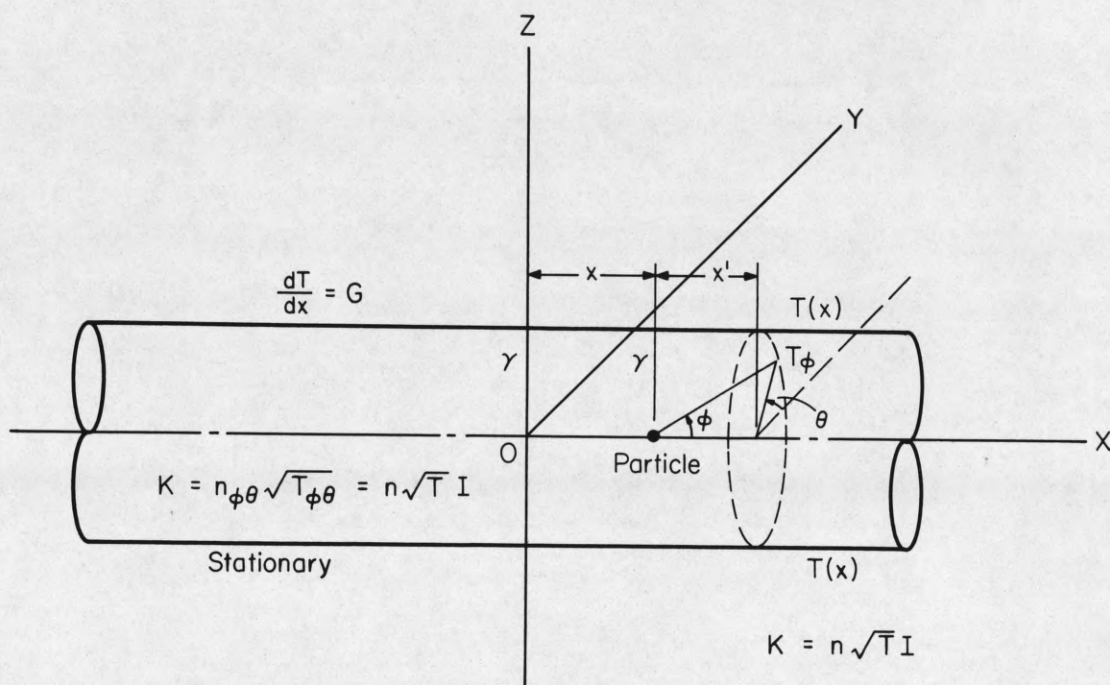
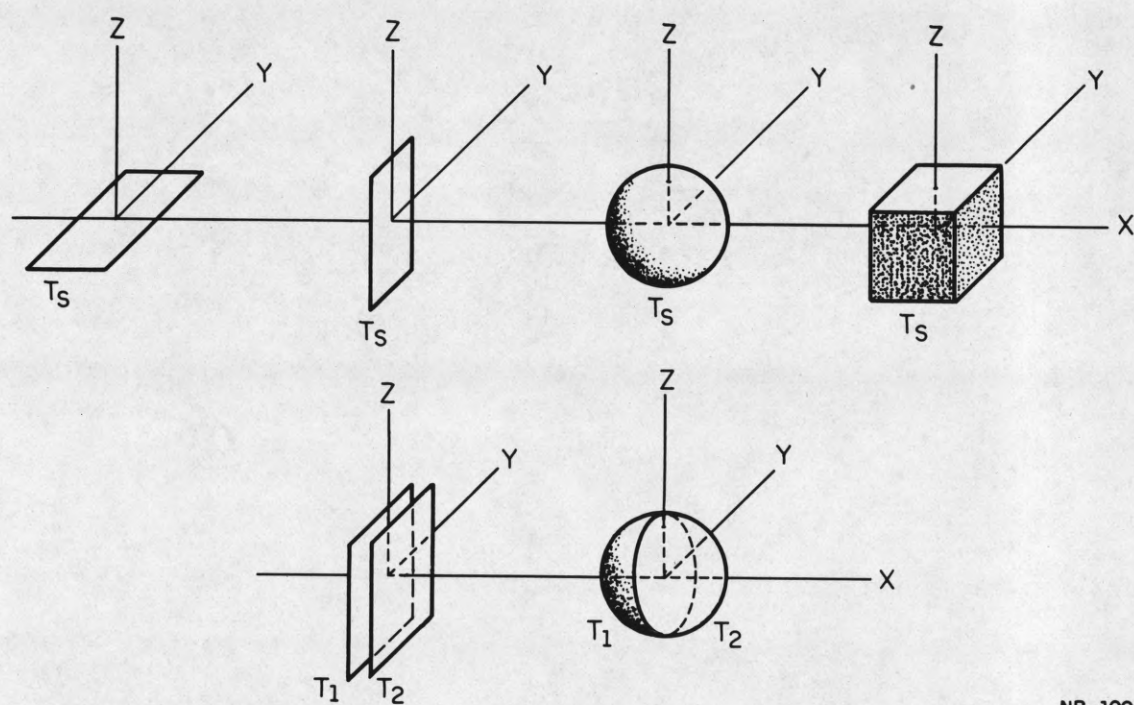


Figure 1. Particle in a Closed System (Knudsen Gas)



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Figure 2. Particle in a Cylinder



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Figure 3. Radiometric Photophoresis

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