

Equations of state of a dilute gas under a heat flux

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Thermal and caloric equations of state for a dilute gas under a heat flux are used to provide a possible microscopic interpretation of temperature in nonequilibrium states. Two possible experiments in which this nonequilibrium temperature could be shown are discussed. They include the study of the Doppler broadening of spectral lines, and of the velocity of long sound waves, both in a nonequilibrium situation characterized by a steady heat flux.

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I. INTRODUCTION

Recent theories in nonequilibrium thermodynamics [1–5] have proposed the use of a generalized nonequilibrium entropy depending not only on the classical variables but also on the dissipative fluxes, as, for instance, the heat flux. These so-called extended thermodynamic theories yield for a monatomic ideal gas, up to second order in the heat flux \mathbf{q} , an entropy of the form

$$s(u, v, \mathbf{J}) = s_{\text{eq}}(u, v) - \frac{\tau \rho}{2\lambda T^2} \mathbf{J} \cdot \mathbf{J}. \quad (1)$$

Here, s is the specific nonequilibrium entropy, s_{eq} the specific local-equilibrium entropy, u and $v = \rho^{-1}$ are the specific internal energy and specific volume, respectively, T the local-equilibrium absolute temperature, λ the thermal conductivity, τ the relaxation time of the heat flux \mathbf{q} , and $\mathbf{J} = v\mathbf{q}$. We have written the entropy function in terms of \mathbf{J} rather than of \mathbf{q} because, as it is well known in classical thermodynamics, the entropy must be expressed in terms of extensive variables in order to be a thermodynamic potential.

Some comments are in order referring to Eq. (1). It must be taken into account that $s(u, v, \mathbf{J})$ is a phenomenological macroscopic function which coincides with the Boltzmann entropy of the kinetic theory of gases in the truncated form limited to the thirteen-moment development proposed by Grad [6, 7]. Some authors have proposed calling the phenomenological $s(u, v, \mathbf{J})$ a quasientropy, or calortropy, to emphasize the difference between the microscopic Boltzmann entropy.

The differential of (1) may be written as

$$ds = \theta^{-1} du + \pi \theta^{-1} dv - \frac{\tau \rho}{\lambda T^2} \mathbf{J} \cdot d\mathbf{J}, \quad (2)$$

where θ and π are given by

$$\theta^{-1} = \left(\frac{\partial s}{\partial u} \right)_{v, \mathbf{J}}, \quad \pi \theta^{-1} = \left(\frac{\partial s}{\partial v} \right)_{u, \mathbf{J}}. \quad (3)$$

In local equilibrium, i.e., in the limit when $\tau \rightarrow 0$, θ and π coincide, respectively, with T and p , the local-equilibrium expressions for the absolute temperature and pressure. Some researchers have proposed that θ and π are the absolute temperature and pressure which are actually mea-

sured in nonequilibrium states instead of T and p , which are defined by the usual expressions $(3/2)kT/m = u$ and $p = nkT$, k being the Boltzmann constant, m the mass of a molecule, and n the number of particles per unit volume.

Other authors, instead, do not attribute to θ and π a real physical meaning except in local equilibrium. In any case, in spite of the lengthy and subtle arguments on the meaning of these quantities [8, 9], the final answer should come from experience. Here, we examine two possible experiments which could shed some light on this subtle topic and which complement a previous more abstract proposal [8a,b]. In fact, the present analysis, which is centrally concerned with the proposal of experiments, will also illustrate the subtleties behind this difficult subject. The physical situation to be considered consists of a filled tank gas placed between two heat reservoirs at different temperatures, so that a constant heat flux q_y is maintained through the system (see Fig. 1).

Some aspects of this problem could probably be dealt with by starting from the Boltzmann equation. The problem is, however, that there are several different methods to solve the Boltzmann equation, and that there are several different boundary conditions compatible with given macroscopic information. Thus a macroscopic analysis of some of the questions arising in the context of second-order transport theories could be useful to obtain new criteria to choose among the different results obtained from the Boltzmann equation by different methods.

The plan of the paper is as follows. In Sec. II, the thermal and caloric equations of state for a monatomic dilute gas under a stationary heat flux are obtained, their deviations with respect to the classical local-equilibrium results underlined, and a microscopic interpretation of the generalized temperature is given. Section III analyzes two possible experiments which could show the consequences of the generalized temperature; on the one hand, we study the Doppler broadening of spectral lines in a dilute gas under a heat flux and, on the other, we show the modifications that extended thermodynamics introduces in the speed of a low-frequency sound wave traveling perpendicular to the temperature gradient. Finally, Sec. IV is devoted to some concluding remarks.

II. EQUATIONS OF STATE OF A DILUTE GAS UNDER A HEAT FLUX

When one takes into account that for monatomic ideal gases $\lambda = \frac{5}{2}(k^2 T n/m)\tau$, one may obtain from (1) the following explicit expression for the specific entropy of the gas,

$$s(u, v, \mathbf{J}) = s_{\text{eq}}(u, v) - \frac{27}{40} \frac{k}{m u^3} \mathbf{J} \cdot \mathbf{J}. \quad (4)$$

Thus, one finds for the generalized temperature

$$\theta^{-1} = \left(\frac{\partial s}{\partial u} \right)_{v, \mathbf{J}} = \frac{1}{T} + \frac{81}{40} \frac{k}{m u^4} \mathbf{J} \cdot \mathbf{J}, \quad (5)$$

and for the thermal equation of state

$$\frac{\pi}{\theta} = \left(\frac{\partial s}{\partial v} \right)_{u, \mathbf{J}} = \frac{p}{T} = nk, \quad (6)$$

since the nonequilibrium part in (4) yields a vanishing contribution. Therefore, it turns out that with this definition of pressure the thermal equation of state for ideal gases has the same form as in equilibrium, but with π and θ instead of p and T . Note, of course, that Eq. (6) does not imply that $p = \pi$ and $T = \theta$.

Relation (6) allows a simple microscopic interpretation for the nonequilibrium temperature. The pressure tensor under a heat flux takes the form [9]

$$\mathbf{P} = \pi \mathbf{U} + \alpha \mathbf{q} \mathbf{q}, \quad (7)$$

with \mathbf{U} the unit tensor and α a scalar quantity satisfying the relation $\text{tr} \mathbf{P} = 3p$, as imposed by the microscopic expressions for the local-equilibrium temperature, $u = \frac{3}{2} kT/m$, and for the pressure tensor \mathbf{P} , namely

$$P_{ij} = \int m v_i v_j f(\mathbf{v}) d\mathbf{v}, \quad (8)$$

where f denotes the nonequilibrium distribution function and \mathbf{v} the molecular velocity. If one assumes that the heat flux is in the y direction, one has, according to (6)–(8),

$$P_{xx} = \pi = nk\theta = nm \langle v_x^2 \rangle_{\text{neq}}, \quad (9)$$

with $\langle \cdot \rangle_{\text{neq}}$ standing for the average in the nonequilibrium steady state. Therefore, the nonequilibrium temperature θ may be interpreted as

$$\theta = \frac{m}{k} \langle v_x^2 \rangle_{\text{neq}} = \frac{m}{k} \langle v_z^2 \rangle_{\text{neq}} \neq \frac{m}{k} \langle v_y^2 \rangle_{\text{neq}}. \quad (10)$$

This relation shows a breakdown of the equipartition law in the presence of a heat flux.

On the other side, one defines the local-equilibrium temperature T by its usual relation with the mean kinetic translational energy of the particles as [6]

$$T = \frac{1}{3} \frac{m}{k} \langle v_x^2 + v_y^2 + v_z^2 \rangle_{\text{neq}}. \quad (11)$$

Then, one could use kinetic temperatures $\theta_x = (m/k) \langle v_x^2 \rangle$, $\theta_z = (m/k) \langle v_z^2 \rangle$, and $\theta_y = (m/k) \langle v_y^2 \rangle$, and

one finds from (5) that $\theta_x = \theta_z = \theta < T$, and from (11) that $\theta_y = 3T - 2\theta > T$. Note that (9), (10), and (11) do not depend on the explicit expression of the microscopic distribution function.

Thus, the presence of a heat flux has some similarities with the presence of a magnetic field \mathbf{B} in producing an anisotropy in the system implying different kinetic temperatures along \mathbf{B} and perpendicular to \mathbf{B} . The effect of the magnetic field, however, is dynamical, since it exerts a force on moving particles, whereas the influence of \mathbf{q} is purely statistical.

Equation (7) predicts different results for the pressure measured in the x and y directions in the nonequilibrium steady state. According to (7), this difference amounts to $\alpha \mathbf{q}^2$. In order to express this value in terms of directly measurable quantities, one uses the condition $\text{tr} \mathbf{P} = 3p$ and the thermal equation of state (6) to obtain

$$P_{yy} - P_{xx} = \alpha \mathbf{q}^2 = 3nk(T - \theta). \quad (12)$$

Finally, with the help of the caloric equation of state (5), the pressure difference writes

$$P_{yy} - P_{xx} = \frac{6}{5} \frac{m}{n^2 k^3 \theta^3} \mathbf{q}^2 = \frac{6}{5} \frac{nm}{\pi^3} \mathbf{q}^2. \quad (13)$$

Then, for measures taken at the same height, extended thermodynamics predicts that the pressure in the y direction is higher than in the x and z directions, and consequently the work required for a volume change is smaller if the force is applied in the x or z directions than in the y direction.

Let us also notice that our thermodynamic pressure π corresponds to the minimum eigenvalue of the pressure tensor \mathbf{P} . This is in agreement with the conjecture raised by Evans in view of the results of his very extensive and systematic numerical simulations of systems in steady states, which he justified by pointing out that “if the entropy is related to the minimum reversible work required to accomplish a virtual volume change in a nonequilibrium steady state, then πdV is the minimum pV work that is possible” [10].

III. TWO POSSIBLE EXPERIMENTS

The aim of this section is to draw the general lines for two possible experiments which could show the differences between θ and T .

A. Doppler broadening of spectral lines

The anisotropy in the pressure tensor could be detected by careful dynamical analyses of the forces acting along the heat flux direction or perpendicular to it. However, there is still a better method which could be much more sensitive to the anisotropy in the second moments of the velocity. Such a method would consist in exciting, by means of two crossed laser beams, a small region of the fluid, and in measuring the width of the spectral lines of the relaxation signal along the direction of the heat flux and perpendicular to it. According to our predictions, the Doppler broadening of the lines (which would

be observable at low pressures, in a domain consistent with our dilute gas limit) should be less in the direction perpendicular to the heat flux than along the heat flux, because the former is proportional to $\langle v_x^2 \rangle$ and the latter to $\langle v_y^2 \rangle$, whose expressions are given after (11). More explicitly, one would have for the width of the spectral lines measured along the x and y directions [11]

$$(\Delta\nu)_x = \frac{\nu_0}{c} \sqrt{\frac{k\theta}{m}} < \frac{\nu_0}{c} \sqrt{\frac{kT}{m}}, \quad (14)$$

$$(\Delta\nu)_y = \frac{\nu_0}{c} \sqrt{\frac{k(3T - 2\theta)}{m}} > \frac{\nu_0}{c} \sqrt{\frac{kT}{m}}, \quad (15)$$

with $(\Delta\nu)$ the root mean square of the intensity distribution, ν_0 the frequency of the light emitted by a molecule at rest, and c the light speed.

This difference between $\langle v_x^2 \rangle$ and $\langle v_y^2 \rangle$ may be understood from a microscopic point of view by considering the following form for the steady state distribution function [9]:

$$f = A \exp \left[-\beta \frac{1}{2} m v^2 \right] \left[1 - \gamma \cdot \left(\frac{1}{2} m v^2 - \frac{5}{2\beta} \right) \mathbf{v} + \frac{(\gamma \cdot \mathbf{v})^2}{2} \left(\frac{1}{2} m v^2 - \frac{5}{2\beta} \right)^2 \right], \quad (16)$$

with \mathbf{v} the peculiar velocity of the gas molecules. Such a distribution function is the second-order development of the maximum entropy distribution function of the form

$$f = A \exp \left[-\beta \frac{1}{2} m v^2 - \gamma \cdot \left(\frac{1}{2} m v^2 - \frac{5}{2\beta} \right) \mathbf{v} \right], \quad (17)$$

where β and γ are Lagrange multipliers related to the mean energy and to the mean heat flux, and are determined from

$$\int f \frac{1}{2} m v^2 d\mathbf{v} = \frac{3}{2} n k T,$$

$$\int f \frac{1}{2} m v^2 \mathbf{v} d\mathbf{v} = \mathbf{q},$$

and A is a normalization constant. This form of the distribution function has been discussed explicitly in [9], so that we will not comment on it here with more detail. It must be noted that up to first order, one recovers the Grad distribution function. However, the higher-order Grad analyses do not predict any change in the diagonal second moments $\langle v_x^2 \rangle$, $\langle v_y^2 \rangle$, and $\langle v_z^2 \rangle$ because they use for the distribution function a development of the form

$$f = A \exp \left[-\frac{m v^2}{2kT} \right] \left[1 + \sum_{l=1}^{\infty} a^{(l)}(\mathbf{r}, t) : H^l(\mathbf{v}) \right], \quad (18)$$

with $H^l(\mathbf{v})$ orthonormalized Hermite polynomials of the velocity whose contribution to $\langle v_x^2 \rangle$, $\langle v_y^2 \rangle$, and $\langle v_z^2 \rangle$ are null, due to the orthogonality conditions. Therefore, in

Grad's expansion β is always equal to $1/kT$, with T the local equilibrium temperature, and $\langle v_x^2 \rangle = \langle v_y^2 \rangle = \langle v_z^2 \rangle = kT/m$, in contrast to what happens in (16).

To go deeper inside the differences between (16) and (18), note that one may assume different boundary conditions on the wall, and that the Boltzmann equation by itself cannot decide which boundary conditions are the most satisfactory ones. For instance, a prescribed heat flux \mathbf{q} may be achieved with very different prescriptions for higher-order moments of f . Indeed, in the Grad thirteen-moment expansion one assumes that the mean values of $H^l(\mathbf{v})$ for $l > 3$ vanish identically. However, one could have the same value of \mathbf{q} but with many different possibilities concerning the mean values of $H^l(\mathbf{v})$, $l > 3$. Thus, the prescription of given values for the mean internal energy and mean heat flux are not enough to single out one specific solution for the distribution function. The distribution function (16) is also compatible with the given mean values of the energy and the heat flux, but it yields nonvanishing mean values for $H^l(\mathbf{v})$, ($l > 3$). Both (16) and (18) could be introduced into the Boltzmann equation to obtain the evolution equations for $\gamma(\mathbf{r}, t)$ as for the set $a^{(l)}(\mathbf{r}, t)$. Which of (16) or (18) gives a better description of the actual situation thus requires an analysis of second-order terms in the equations of state, which is one of the differences that follows from (16) and (18). Another possibility would be to measure the higher-order moments of the distribution function. Here we have emphasized the differences in the equations of state because they have received almost no attention as compared with the analysis of higher-order transport equations.

B. Sound speed in a dilute gas under a heat flux

In this section, we analyze how the nonequilibrium corrections to the equations of state affect the speed of a sound wave propagating perpendicular to the heat flux—i.e., in the x direction—thus pointing out another experimental test of the theory by measuring the velocity of a sound wave under nonequilibrium stationary situations.

The experimental situation should be as follows (Fig. 1). A tank containing a monatomic gas is placed be-

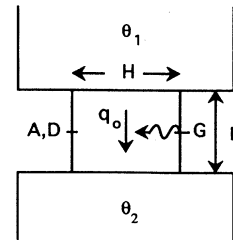


FIG. 1. Picture of the physical situation. A steady flux q_0 is maintained through a gas container by keeping it in contact with two heat reservoirs at different temperatures θ_1 and θ_2 . A sound generator G , a sound detector D , and a thermometer A are placed at the same height. The sound wave travels perpendicular to the heat flux.

tween two heat baths at different temperatures, so that the system reaches a steady situation characterized by vertical temperature and density profiles, and a constant heat flux \mathbf{q}_0 crossing the whole system. Obviously, the hot reservoir is placed at the top and the cold one at the bottom, in order to prevent any convective instability. A thermometer, a sound generator, and a detector are placed at the same height y_0 . Our purpose is to evaluate the velocity of a sound wave which travels along this horizontal layer. To this end, we proceed as usual by linearizing the mass and momentum balance equations

$$\frac{d\rho}{dt} + \rho \nabla \cdot \mathbf{v} = 0, \quad (19)$$

$$\rho \frac{d\mathbf{v}}{dt} + \nabla \cdot \mathbf{P} = \mathbf{0} \quad (20)$$

around the stationary state characterized by $\rho = \rho_0(y)$, \mathbf{q}_0 , and $\mathbf{v} = \mathbf{0}$.

For long wavelengths, we will consider, as usual, that gradients in the direction of wave propagation are small enough as to neglect dissipative effects, i.e., viscous effects are negligible and \mathbf{q}' , the perturbation to the heat flux due to the sound wave, is small compared to \mathbf{q}_0 , that is to say, $|\mathbf{q}'| \ll |\mathbf{q}_0|$. Then, the pressure tensor \mathbf{P} has no contribution due to viscosity and reduces to expression (7). It is immediate to find that a perturbation propagating along the x direction satisfies a wave equation with velocity $c = (\partial\pi/\partial\rho)_{s,\mathbf{q}}^{1/2}$. The latter derivative is taken at constant s and \mathbf{q} since it is assumed that the process is carried out without net heat exchange, and that the heat flux deviations with respect to the stationary value \mathbf{q}_0 are negligible according to the long wavelength condition. To assume $\mathbf{q}_0 \neq \mathbf{0}$ is not contradictory with the assumption that the net heat exchange during the process is zero, because \mathbf{q}_0 is constant along the y direction so that the heat input is equal to the heat output.

To evaluate the partial derivative, we start from the condition of vanishing heat exchange. Since π is the actual pressure in the nonequilibrium state, one has for the differential work in an infinitesimal volume change $dW = -\pi dv$, so that the corresponding expression for the heat exchange yields

$$dQ = du + \pi dv = 0. \quad (21)$$

The fact that $dQ = 0$ does not exactly imply that $ds = 0$ in (2); however, the contributions of \mathbf{q}_0 to π are of the order \mathbf{q}_0^2 , whereas the contribution to ds of the last term in (2) is of the order $\mathbf{q}_0 \cdot \mathbf{q}'$, i.e., much smaller than $|\mathbf{q}_0^2|$. Therefore, the exact relationship between heat and the generalized entropy only plays an important role in the equations for θ and π , which have been given in (5) and (6), but it has a negligible influence on the expression for the sound speed.

In order to evaluate the differential of energy, we use the equations of state (5) and (6). By using that $T = 2mu/3k$ for a monatomic gas, Eq. (5) yields for the specific internal energy, up to order \mathbf{q}_0^2 ,

$$u = \frac{3}{2} \frac{k}{m} \theta \left[1 + \frac{2}{5} \frac{m^3 v^2}{k^3 \theta^3} \mathbf{q}_0^2 \right] = \frac{3}{2} \pi v + \frac{3}{5} \frac{1}{\pi^2} \mathbf{q}_0^2, \quad (22)$$

the last equality coming from Eq. (6). Finally, by differentiating (22), Eq. (21) leads for the sound speed, after some straightforward calculations up to order \mathbf{q}_0^2 ,

$$c = \left(\frac{\partial\pi}{\partial\rho} \right)_{s,\mathbf{q}} = \sqrt{\frac{5k\theta}{3m}} \left(1 + \frac{2}{5} \frac{m}{k\theta\pi^2} \mathbf{q}_0^2 \right) \equiv c_0(1 + \Delta), \quad (23)$$

where we have denoted with c_0 the local-equilibrium sound speed, and with Δ the nonequilibrium correction. Equation (23) predicts a modification in the sound velocity for the gas in a steady nonequilibrium situation with respect to the local-equilibrium result. Let us now examine under which experimental conditions these corrections could be experimentally attainable.

Let us first notice that the deviation from the local-equilibrium result Δ can be written in a simpler way by using the microscopic expression for the thermal conductivity of a monatomic gas. Direct calculations yield, aside from a factor of order unity,

$$\Delta \simeq \left(l \frac{\nabla\theta}{\theta} \right)^2, \quad (24)$$

with l the mean free path of the molecules. The temperature gradient can be obtained by using the fact that in the stationary state, \mathbf{q}_0 is uniform over the system. Since the thermal conductivity for a monatomic gas depends on temperature as $\lambda \propto \theta^{1/2}$ [11], one has

$$\nabla\theta^{3/2} = \text{const} = \frac{\theta_1^{3/2} - \theta_2^{3/2}}{L}, \quad (25)$$

θ_1 and θ_2 being the temperatures of the hot and cold reservoirs, respectively, and L is the separation between them, that is to say, the height of the sample. Therefore, by introducing (25) into (24), the Knudsen number K satisfies the relation

$$K = \frac{l}{L} \simeq \Delta^{1/2} \frac{\theta_0^{3/2}}{\theta_1^{3/2} - \theta_2^{3/2}}. \quad (26)$$

For the regime to be diffusive, as we are considering in (25), it must be satisfied that $l/L \ll 1$. This imposes some restrictions on the value of the temperature difference between the reservoirs, and on the temperature θ_0 of the layer where the sound wave is traveling. In particular, it is concluded that the temperature difference should be large, as it must be in the conditions of the experiment, and that the temperature at which the sound speed is measured θ_0 must be small. In order to establish how small this temperature should be, by taking $\theta_1 \gg \theta_2$, Eq. (26) can be rearranged to give

$$\frac{\theta_0}{\theta_1} \simeq \frac{K^{2/3}}{\Delta^{1/3}}. \quad (27)$$

If one requires $K < 10^{-2}$ and $\Delta = 10^{-2}$, that is to say, for a nonequilibrium correction of the order of 1%, relation (27) implies $\theta_0 < 0.2\theta_1$.

Then, if one takes for the temperatures of the baths $\theta_1 = 1000$ K and $\theta_2 = 200$ K, and for the measuring temperature $\theta_0 = 250$ K, the Knudsen number amounts

to $l/L = 1.2 \times 10^{-2}$, so that for a typical value $L = 10$ cm, one has $l \simeq 1$ mm. The previous value for the mean free path determines the density of the gas in the layer where the measure must be taken; indeed the expression $l = 1/n\pi d^2$, with d the molecule diameter (π here is the π number, not to be confused with the generalized pressure), implies that for a typical value of $d = 4$ Å, the number density in order that $l \simeq 1$ mm is $n \simeq 6 \times 10^{21}$ m⁻³, and the corresponding generalized pressure $\pi = nk\theta$, for the temperature $\theta_0 = 250$ K adopted above, amounts to $\pi \simeq 20$ Pa. Let us remark that, in contrast to the temperature or the density, which depend on the height y , the pressure is uniform over the system due to the stationary condition with vanishing barycentric speed.

On the other hand, in order to be qualified to treat the medium as a continuum, as we did in the calculation of the sound wave, it is necessary that the length of the system in the x direction H be much larger than the mean free path. This constrains the value of H to a minimum of, say, 10 cm, for the values of the parameters taken before.

IV. CONCLUDING REMARKS

The analysis of temperature in nonequilibrium situations is subtle and complex, and it is difficult to propose experiments to settle this conflictive but fundamental topic. Therefore, we feel that a theoretical effort is needed to propose different experiments, whose practical realization would require, of course, much care. In this paper, we have proposed two such experiments more realistic than those proposed up to now. Furthermore, we have seen that the nonequilibrium temperature θ is not contradictory with the definition of a local-equilibrium temperature T related to the mean energy. The problem is, of course, whether θ or T is measured in a given

experiment.

We have seen that the nonequilibrium temperature θ could have a kinetic definition in terms of $\langle v_x^2 \rangle_{\text{neq}}$, but that this does not imply that θ is equal to T , which is related to $\langle v_x^2 + v_y^2 + v_z^2 \rangle_{\text{neq}}$. The anisotropy due to the presence of the heat flux \mathbf{q} causes the second moments of the velocity distribution to be such that $\langle v_x^2 \rangle_{\text{neq}} \neq \langle v_x^2 + v_y^2 + v_z^2 \rangle_{\text{neq}}/3$, the first average related to θ , and the second one to T , thus breaking the equipartition of energy. Having a kinetic interpretation of θ and T is useful for numerical simulations, in which such averages can be easily obtained.

The relation of the generalized temperature with the second moments of the velocity distribution is also interesting from the point of view of a parallelism between our result and those of Keizer [12]. Indeed, the latter author has proposed measuring nonequilibrium temperatures by evaluating the second moments of the fluctuations of macroscopic quantities. However, he has not considered the effect of the relaxation time in heat transport in fluids, where he has always used Fourier's law. As a consequence, he has not considered the situation dealt with in this paper.

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