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ON THE BEHAVIOUR OF A RELAXING GAS IN A CYLINDRICAL HEAT-CONDUCTIVITY CELL

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Abstract—The behaviour of a relaxing gas in a cylindrical heat-conductivity cell is re-assessed and attention drawn to some special circumstances which do not appear to have been noted before. Firstly, it is shown that the homogeneous gas phase can be maintained in thermal equilibrium (regardless of the effectiveness of homogeneous excitation) by surface effects alone. Secondly, it is shown that a rapidly relaxing gas may exhibit all the characteristics of a monatomic gas in respect of the dependence of measured conductivity on pressure.

NOMENCLATURE

a_n, b_n $(n = 1, 2)$,	functions defined in equation (10);
c,	specific heat of relaxing internal energy mode;
$\mathscr{D}$,	diffusion coefficient;
I_0, K_0,	modified Bessel functions of order zero;
l_0,	mean free path;
L,	Lewis–Semenov number [equation (7)];
n,	number density;
N,	number of collisions required to excite internal mode;
P_1, P_2, Q,	functions in equations (12)–(14);
q, q_1,	energy flux; value at inner cylinder;
r,	radial co-ordinate;
R_1, R_2,	values of r at inner and outer cylinders;
T,	translational temperature;
T_{w1}, T_{w2},	temperatures of cylindrical surfaces at R_1 and R_2;
α, β,	defined in equation (6);
ϑ,	internal mode temperature;
λ,	translational conductivity;
λ_a,	measured conductivity;
λ_E,	Eucken-corrected conductivity;
τ,	relaxation time;
ϕ,	defined in equation (22).

1. INTRODUCTION

THERE have been a number of theoretical assessments of the behaviour of relaxing or reacting gases in heat-conductivity cells, the latter usually consisting of either parallel (infinite) flat plates or of pairs of concentric circular cylinders. In what follows we shall confine our remarks to the case of relaxing internal energy modes.

In order to estimate the steady flux of energy across the cell it is necessary to know how the internal mode excitation takes place in the homogeneous phase and, in addition, how both the translational (or translational-plus-active) and relaxing modes are excited or de-excited on collision with the walls. In other words it is necessary to know the accommodation coefficients for both modes of energy storage. This set of boundary conditions makes the algebra involved in the solution rather heavy, and the direct physical interpretation of the eventual results somewhat awkward. The present paper attempts to simplify the presentation of the final solution as much as possible and, in doing so, points to some special circumstances which may be of some interest and which appear to have been overlooked by previous investigators.

2. THE GENERAL SOLUTION

For the sake of completeness we shall begin with a brief derivation of the basic equations. An exhaustive account is not necessary since they have been adequately discussed for a wide range of circumstances in, for example, references 1–5. We shall consider the case of cylindrical symmetry so that, under the steady state conditions, the energy equation gives

$$rq = \text{constant}. \tag{1}$$

Writing T for the temperature of the translational-plus-active modes, and assuming that the energy content of the relaxing mode can be specified in terms of a temperature ϑ, it can be shown (e.g. [4]) that

$$q = -\lambda\frac{dT}{dr} - n\mathscr{D}c\frac{d\vartheta}{dr}. \tag{2}$$

Evaluating the constant in equation 1 at the surface of the inner cylinder, where $r = R_1$ and $q = q_1$, leads to the result that

$$r\lambda\frac{dT}{dr} + rn\mathscr{D}c\frac{d\vartheta}{dr} = -R_1q_1. \tag{3}$$

The relaxation equation, which expresses the balance between diffusive efflux from and excitation of the internal mode in a volume element, can be written [4] in the form

$$\frac{d}{dr}\left(rn\mathscr{D}c\frac{d\vartheta}{dr}\right) = \frac{rn}{\tau}\int_T^{\vartheta} c(\vartheta')\,d\vartheta', \tag{4}$$

where τ is the relevant relaxation time. With the knowledge that the pressure must be constant throughout the cell, so that the number density n, the translational conductivity λ and the diffusion coefficient $\mathscr{D}$ are all functions of T, equations (3) and (4) form a complete set from which T and ϑ can, in principle anyway, be determined. (N.B. We imply that c, the specific heat of the relaxing mode is also a known function of ϑ. In addition, in deriving equations (2) and (4), the assumption has been made that the diffusion coefficient $\mathscr{D}$ is the same for all molecules, regardless of their internal quantum state; this is a reasonable approximation provided that the molecules remain in their electronic ground state.)

It is quite justifiable for present purposes to simplify equations (3) and (4) by evaluating all of n, λ, $\mathscr{D}$, c and τ at some suitable mean cell temperature, and indeed this assumption has been made by all previous investigators. Practical temperature differences across a cell are invariably small enough to warrant this assumption. Denoting such average values by adding a subscript 0 it can easily be shown that

$$r\frac{d}{dr}\left(\frac{1}{r}\frac{d\psi}{dr}\right) - \alpha^2\psi = \alpha^2\beta, \tag{5}$$

where ψ is either $r(dT/dr)$ or $r(d\vartheta/dr)$. The constants α and β are defined so that

$$\alpha^2 = \frac{1+L}{\tau_0\mathscr{D}_0};\quad \beta = \frac{R_1q_1}{\lambda_0(1+L)}, \tag{6}$$

where L is a Lewis–Semenov number, given by

$$L = \frac{n_0\mathscr{D}_0c_0}{\lambda_0}. \tag{7}$$

Equation 5 can be solved to show that

$$T = AI_0(\alpha r) + BK_0(\alpha r) - \beta\log r + C. \tag{8}$$

I_0 and K_0 are the zero-th order modified Bessel functions [6] and A, B and C are constants to be determined. The solution for ϑ is of similar form to equation (8) with constants which, in general, differ from A, B and C. However, the appropriate forms of equations (3) and (4), together with equations (6) and (7), show that

$$\vartheta = -\frac{1}{L}\{AI_0(\alpha r) + BK_0(\alpha r)\} - \beta\log r + C. \tag{9}$$

Similar results have either been obtained or are implied in references 1–3 and 5, for example.

In any practical problem, the temperatures of the solid cylindrical surfaces can be considered as given quantities; we shall write T_{w1} for the temperature of the surface $r = R_1$ and T_{w2} for the temperature at $r = R_2$. (N.B. $R_2 > R_1$.) Then we shall have four boundary conditions which, taking account of the temperature jumps at R_1 and R_2, can be written as

$$\left.\begin{aligned} T - T_{w1} &= a_1\left(\frac{dT}{dr}\right); & r &= R_1,\\ T - T_{w2} &= -a_2\left(\frac{dT}{dr}\right); & r &= R_2,\\ \vartheta - T_{w1} &= b_1\left(\frac{d\vartheta}{dr}\right); & r &= R_1,\\ \vartheta - T_{w2} &= -b_2\left(\frac{d\vartheta}{dr}\right); & r &= R_2, \end{aligned}\right\} \tag{10}$$

The four quantities a_1, a_2, b_1 and b_2 are all positive and are directly related to the accommodation coefficients for translational-plus-active energy (a_1, a_2) and for the relaxing mode energy (b_1, b_2), and also to the free paths for transfer of the particular form of energy involved [4]. There is no need for us to write down the explicit forms of a_n and b_n, but we may note that they are all, in general, inversely proportional to the pressure. The four conditions just given are necessary because β in equations (8) or (9) must also be considered as an unknown constant. The reason is that q_1 is, initially, unknown and will be determined by the physical constants pertaining to the gas and cell in question once T_{w1} and T_{w2} are specified.

The two conditions at $r = R_2$ are replaced in reference 1 by the requirement that $\vartheta \to T$ as $r \to \infty$, so that Schäfer, Rating and Eucken have really considered the problem of an isolated wire in an infinite expanse of gas which is, asymptotically, in an equilibrium state. When R_2 is greater than R_1 by a sufficient amount, this approximation to the concentric cylinder case may be adequate, but the complete solution for finite R_2 is not prohibitively complicated and indeed has been given by other writers since the paper by Schäfer, Rating and Eucken [1] appeared. One example of this latter solution has been given by Wright [3]. However this solution is complicated by the fact that Wright is interested in relating a measured conductivity to two gas temperatures detected at positions r_{I} and r_{II} such that $R_1 < r_{\text{I}} < r_{\text{II}} < R_2$. This extra complication is introduced so as to obtain conductivity measurements in which "... temperature discontinuities at the boundaries no longer interfere." The inference is then drawn that, if apparent (measured) conductivities obtained in this way vary with pressure, such variations will signify the presence of relaxation effects. We make some comments on these matters below.

Confining attention to the constant β, it can be shown after some algebra that

$$\begin{aligned}\beta\left\{\log\left(\frac{R_2}{R_1}\right) + \frac{1}{R_1}\left[a_1 + (b_1 - a_1)\frac{P_1}{Q}\right]\right.&\\ \left. + \frac{1}{R_2}\left[a_2 + (b_2 - a_2)\frac{P_2}{Q}\right]\right\} &= T_{w1} - T_{w2}, \quad (11)\end{aligned}$$

where

$$\left.\begin{aligned}P_1 &= L(1 + L)W_1 + L\alpha(b_2 + La_2)\\ &\quad [(\alpha R_2)^{-1} - W_3]\\ &\quad - L(1 + L)[a_2R_2^{-1} + \alpha a_1W_2]\\ &\quad + L\alpha^2a_1(b_2 + La_2)W_4,\end{aligned}\right\} \quad (12)$$

$$\left.\begin{aligned}P_2 &= L(1 + L)W_1 + L\alpha(b_1 + La_1)\\ &\quad [(\alpha R_1)^{-1} - W_2] - L(1 + L)\\ &\quad [a_1R_1^{-1} + a_2W_3] + L\alpha^2a_2\\ &\quad (b_1 + La_1)W_4,\end{aligned}\right\} \quad (13)$$

$$\left.\begin{aligned}Q &= (1 + L)^2W_1 - (1 + L)\\ &\quad [\alpha(b_1 + La_1)W_2 + \alpha(b_2 + La_2)W_3]\\ &\quad + \alpha^2(b_1 + La_1)(b_2 + La_2)W_4.\end{aligned}\right\} \quad (14)$$

The quantities W_1 to W_4 inclusive are groups of Bessel functions as follows,

$$W_1 = I_0(\alpha R_1)K_0(\alpha R_2) - I_0(\alpha R_2)K_0(\alpha R_1), \quad (15)$$

$$W_2 = I_1(\alpha R_1)K_0(\alpha R_2) + I_0(\alpha R_2)K_1(\alpha R_1), \quad (16)$$

$$W_3 = I_1(\alpha R_2)K_0(\alpha R_1) + I_0(\alpha R_1)K_1(\alpha R_2), \quad (17)$$

$$W_4 = I_1(\alpha R_1)K_1(\alpha R_2) - I_1(\alpha R_2)K_1(\alpha R_1). \quad (18)$$

If the quantities λ_a and λ_E are now defined so that

$$\begin{aligned}\lambda_a = \frac{R_1q_1}{(T_{w1} - T_{w2})}\log\left(\frac{R_2}{R_1}\right) &=\\ \frac{\lambda_E\beta}{(T_{w1} - T_{w2})}\log\left(\frac{R_2}{R_1}\right)&, \quad (19)\end{aligned}$$

$$\lambda_E = \lambda_0(1 + L), \quad (20)$$

it is possible to re-write equation (11) in the form

$$\frac{1}{\lambda_a} = \frac{1}{\lambda_E} + \phi, \quad (21)$$

where

$$\begin{aligned}\phi = \left[\lambda_E\log\left(\frac{R_2}{R_1}\right)\right]^{-1}&\left[\frac{1}{R_1}\left\{a_1 + (b_1 - a_1)\frac{P_1}{Q}\right\}\right.\\ &\left. + \frac{1}{R_2}\left\{a_2 + (b_2 - a_2)\frac{P_2}{Q}\right\}\right]. \quad (22)\end{aligned}$$

λ_a is the measured, or apparent, conductivity which can be deduced from any given single experiment. λ_E is the Eucken-corrected or equilibrium conductivity, as is clear from equations (2) and (7) if we put ϑ equal to T. (Equation (4) shows that ϑ will be equal to T if $\tau = 0$.)

The extent of the difference between λ_a and λ_E is summarized in the quantity ϕ. This shows that λ_a and λ_E differ because of the surface accommodation effects (a_n, b_n; $n = 1, 2$) and because of internal mode relaxation in the gas phase. This latter effect appears in ϕ through the terms P_1, P_2 and Q. The magnitude of the effect of the various excitation efficiencies is dependent on the scale of the cell (i.e. on R_1 and R_2).

3. INTERPRETATION OF THE RESULTS

The final form of the solution given in equation (21) is remarkably simple, especially bearing in mind that it is (within the framework of the initial assumptions) an exact result. For comparison purposes it should be noted that when the relaxing internal mode carries no communicable energy, so that $L = 0$, equation (21) reduces to

$$\frac{1}{\lambda_a} = \frac{1}{\lambda_0} + \phi_0, \tag{23}$$

where

$$\phi_0 = \left[\lambda_0 \log\left(\frac{R_2}{R_1}\right)\right]^{-1}\left[\frac{a_1}{R_1} + \frac{a_2}{R_2}\right]. \tag{24}$$

We shall refer to this as the monatomic result.

Now λ_E is a fixed function in the sense that it does not vary with pressure, and the same is true of λ_0. That λ_a varies with pressure is an experimentally observed fact; equation (21) shows that this variation is entirely described by the function ϕ. We have remarked before that a_n and b_n vary inversely with the pressure. (Any deviation from this mode of variation must be due to changes of accommodation coefficient with pressure. Such changes are usually reckoned to be small over moderate pressure ranges, but could possibly become significant if p varied over several orders of magnitude.) Since both τ_0 and $\mathscr{D}_0$ depend on p^{-1} it follows [equation (6)] that α is proportional to p. Consequently, the products αa_n and αb_n will, in general, be pressure-independent. However, additional variations of ϕ with p will occur because of the appearance of terms involving αR_1 etc., in P_1, P_2 and Q (see equations (12)–(18), inclusive).

It is most important to notice that *all* relaxation effects are contained in the quantities P_1, P_2 and Q and, what is more, that these terms may vanish from ϕ if $b_1 = a_1$ and $b_2 = a_2$. In practical terms these two requirements simply mean that the effectiveness of internal mode accommodation must compare favourably with that of the translational states (however good or bad this may be). We reiterate that if $b_n = a_n$, $n = 1, 2$, the measured conductivity λ_a is *entirely uninfluenced by relaxation effects*, no matter how large or small the relaxation time may be. This special case appears to have been overlooked by previous investigators. Whilst one may not encounter the case exactly in practice, it may well be that one could find $b_n \simeq a_n$. The relaxation effects, whilst not entirely absent in such a case, would certainly be rendered much less significant.

Direct confirmation of the statement that ϕ reduces to the value $\lambda_0\phi_0/\lambda_E$ when $b_n = a_n$ can be obtained by noting that conditions [10] all combine to give

$$T - \vartheta = a_1\frac{\mathrm{d}(T-\vartheta)}{\mathrm{d}r}\ ; \quad r = R_1, \tag{25}$$

$$T - \vartheta = -a_2\frac{\mathrm{d}(T-\vartheta)}{\mathrm{d}r}\ ; \quad r = R_2. \tag{26}$$

Since [equations (8) and (9)]

$$T - \vartheta = \left(1 + \frac{1}{L}\right)\{AI_0(\alpha r) + BK_0(\alpha r)\}, \tag{27}$$

it follows at once that $A = 0 = B$. Thus, although the relaxation time τ_0 may be very different from zero, the gas behaves as if it was in complete thermal equilibrium, and this condition is brought about entirely by the surface accommodation effects.

It is interesting to examine Wright's suggestion [3] that if temperatures T_{I} and T_{II}, say, are measured at radii r_{I} and r_{II} as defined earlier, then temperature jump effects do not interfere with the conductivity measurement. Under the particular conditions $b_n = a_n$ it is readily found that

$$T_{\mathrm{I}} - T_{\mathrm{II}} = (T_{w1} - T_{w2}) \log\left(\frac{r_{\mathrm{II}}}{r_{\mathrm{I}}}\right) \left[\log\left(\frac{R_2}{R_1}\right) + \frac{a_1}{R_1} + \frac{a_2}{R_2}\right]^{-1}, \qquad (28)$$

which, together with equations (6), (11) and (20), shows that

$$\lambda_E = R_1 q_1 \log(r_{\mathrm{II}}/r_{\mathrm{I}})[T_{\mathrm{I}} - T_{\mathrm{II}}]^{-1}. \qquad (29)$$

In this case, therefore, λ_E can be measured directly without interference from surface accommodation effects. What is more, we observe that there will be no effect of pressure on these measurements. However, it would be wrong to infer that the absence of any variations with pressure implied the absence of a relaxing mode. Equation (29) has been arrived at solely as a result of assumptions made about the surface influences. It is clear from equations (11–22) that surface accommodation efficiencies and relaxation effects are most intimately connected with one another in the conductivity cell. If $b_n \neq a_n$ then a relaxation effect will appear in the measured conductivity, but it will be of a magnitude conditioned by the difference between b_n and a_n. It is in fact much safer to argue that an absence of any pressure-dependence in measurements made using the methods discussed in reference 3 is due to the equality of b_n and a_n, rather than to assert that the presence of such dependence is a direct measure of the relaxation effect.

There is another feature of the general result given in equation (21) which does not seem to have been pointed out previously. The analysis used to derive equation (21) is essentially only valid under continuum conditions. If l_0 is the mean free path this requires that we must only use these results if $R_1 \gg l_0$ and $R_2 - R_1 \gg l_0$. The relaxation time τ_0 and diffusion coefficient $\mathscr{D}_0$ can be written in the forms

$$\tau_0 = N l_0 / \Omega_0; \quad \mathscr{D}_0 \doteq l_0 \Omega_0, \qquad (30)$$

where Ω_0 is the arithmetic mean molecular speed and N the number of collisions required to produce thermal equilibrium. Therefore the magnitude of α is roughly equal to $1/l_0\sqrt{N}$. For rapidly relaxing modes like rotation it follows, as a general rule, that the conditions $\alpha R_1 \gg 1$ and $\alpha(R_2 - R_1) \gg 1$ are satisfied. The asymptotic expansions of the Bessel functions in equations (11)–(22) can then be used and it can be shown that

$$\frac{P_n}{Q} \simeq \frac{L(1 + \alpha a_n)}{1 + \alpha b_n + L(1 + \alpha a_n)}; \quad n = 1, 2. \qquad (31)$$

The significant fact is that the P_n/Q are not pressure-dependent (since αa_n and αb_n are not) and reference to equations (21) and (22) will show that plotting $1/\lambda_a$ versus $1/p$ will still lead to a straight line, exactly as in the true monatomic case [equations (23) and (24)], *even though relaxation effects are present.* One can test this assertion by replotting the results of Waelbroeck and Zuckerbrodt [7] for hydrogen and of Taylor and Johnston [8] for air. In the case of reference 7 it has been necessary to read off the results from a rather small graph, so that some inaccuracies may be present on this account. The results of the replotting are shown in Fig. 1, from which it is apparent that the theoretical surmise made above is borne out in practice.

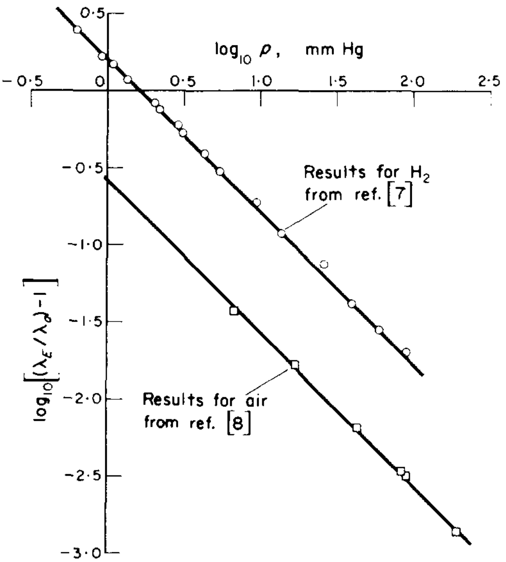

FIG. 1. Experimental results compared with the predictions of equations (21) and (31), namely that

$$\log_{10}\left(\frac{\lambda_E}{\lambda_a} - 1\right) = -\log_{10} p + \log_{10}(p\lambda_E\phi)$$

where $p\lambda_E\phi$ is independent of pressure. The solid lines have slopes of exactly − 45°.

Taylor and Johnston actually remark on the linearity of their $1/\lambda_a$ versus $1/p$ plot in reference 8 and in reference 9 go on to exploit this by calculating accommodation coefficients for several diatomic gases. The theory which they employ to do this does not take account of relaxation effects and gives a result akin to the monatomic solution in equations (23) and (24), with λ_0 being replaced by λ_E. It is reasonable for us to assume [9] that $a_1 = a_2$ and $b_1 = b_2$, so that the slope of their lines $1/\lambda_a$ versus $1/p$ will in fact be equal to

$$\frac{p(1 + R_1/R_2)}{\lambda_E R_1 \log (R_2/R_1)}\left\{ a_1 + \frac{(b_1 - a_1)(1 + \alpha a_1)L}{1 + \alpha b_1 + (1 + a_1)L}\right\} \tag{32}$$

The results of reference 9 are equivalent to the assumption that $b_1 = a_1$, with, however, some allowance being made for the fact that the free path involved in a_1 is modified appropriately (see, for example, reference 4). Itis clear that, even with prior knowledge of the relaxation time (and hence of α) the single measurement leading to the quantity in equation (32) is insufficient to enable one to calculate both a_1 and b_1. It is also important to observe that any temperature variations of the "accommodation coefficients" quoted in reference 9 will include the effects of variation in the relaxation number N with this quantity, a fact which does not seem to have been pointed out before.

Waelbroeck and Zuckerbrodt do not plot $1/\lambda_a$ versus $1/p$; instead they calculate the quantity $(\lambda_a - \lambda_0)/(\lambda_E - \lambda_0)$ from their measurements and plot the result against $\log p$ [7]. The same objection will apply, however, namely that a_1 and b_1 cannot both be calculated from a single measurement, and indeed it is remarked in reference 7 that a sum of rotational and translational accommodation coefficients is obtained from low-pressure experiments and then applied to the reduction of the high-pressure data. That this may not be the most desirable of procedures is evident from the results quoted and discussed in reference 10.

To summarize briefly, the heat-conductivity cell seems to be admirably suited to its primary role, which is the measurement of the conductivity λ_E, but should be used with reservation for the calculation of accommodation effects where rapidly relaxing internal modes are involved.

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Résumé—Le comportement d'un gaz en relaxation dans une cellule cylindrique de mesure de conductivité thermique a été réestimé et l'on s'est intéressé à certaines circonstances spéciales qui ne semblent pas avoir été remarquées auparavant. Tout d'abord, on montre que la phase gazeuse homogène peut être maintenue en équilibre thermique (sans s'occuper de l'efficacité de l'excitation homogène) seulement par des effets de surface. Ensuite, on montre qu'un gaz avec une relaxation rapide peut présenter toutes les caractéristiques d'un gaz monoatomique eu égard à la dépendance de la conductivité mesurée en fonction de la pression.

Zusammenfassung—Das Verhalten eines Gases das sich in einer zylindrischen Wärmeleitzelle entspannt wurde nochmals abgeschätzt und die Aufmerksamkeit auf einige Besonderheiten gelenkt, die bisher nicht bemerkt worden zu sein scheinen. Zuerst wird gezeigt, dass die homogene Gasphase allein durch Oberflächeneffekte in thermischen Gleichgewicht gehalten werden kann (ohne Rücksicht auf die Wirksamkeit der homogenen Anregung). Zweitens wird gezeigt, dass ein sich schnell ausdehnendes Gas hinsichtlich der Druckabhängigkeit der gemessenen Leitfähigkeit alle Charakteristiken eines einatomigen Gases zeigt.

Аннотация—Вновь исследованы характеристики релаксирующего газа в цилиндрической теплопроводящей камере, причем обращено внимание на некоторые особые обстоятельства, которые, оказывается, ранее не были замечены; во-первых, гомогенная газовая фаза может поддерживаться в термическом равновесии (независимо от эффективности гомогенного возмущения) тлоько с помощью поверхностных эффектов, во-вторых, быстро релаксирующий газ может обладать всеми характеристиками одноатомного газа по отношению к зависимости его измеряемой проводимости от давления.