FORMAL RELAXATION OSCILLATIONS FOR A MODEL OF A CATALYTIC PARTICLE*

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I. Introduction. This discussion is motivated by the following system of equations from chemical engineering.

$$\frac{\partial u}{\partial t} = \frac{\partial^2 u}{\partial x^2} - \varphi^2 u F(v), \tag{1}$$

$$\mathrm{Le}\frac{\partial v}{\partial t} = \frac{\partial^2 v}{\partial x^2} + \beta\varphi^2 u F(v) \tag{2}$$

where $F(v) = e^{\gamma(v-1)/v}$. The unknown functions $u(x, t)$ and $v(x, t)$ are proportional to the concentration of reactant and temperature in a one-dimenisonal porous catalytic particle. A complete explanation of this pair of equations can be found in the major treatise of R. Aris [3], whose notation we follow throughout. In particular, the physical significance and observed ranges of the parameters can be found there. (See especially Sec. 2.7). The equations are considered in a region $0 < x < 1$, $t > 0$, with boundary conditions

$$\frac{\partial u}{\partial x} = \frac{\partial v}{\partial x} = 0 \quad \text{at } x = 0, \tag{3}$$

$$\frac{\partial u}{\partial x} = \nu(1 - u) \quad \text{at } x = 1, \tag{4}$$

$$\frac{\partial v}{\partial x} = \mu(1 - v) \quad \text{at } x = 1. \tag{5}$$

The parameters, Le, γ, β, φ, ν and μ are all positive.

Equations (1)–(5) describe a slab of catalyst with Arrhenius kinetics and Robin type boundary conditions. While the particle is considered to occupy the region $-1 \leqslant x \leqslant 1$, only solutions which are symmetric around $x = 0$ will be discussed, resulting in boundary conditions (3)–(5).

A further inspiration for this work is an important numerical study of a variant of this problem which was carried out by J. C. M. Lee and D. Luss [7]. Spherical geometry and Dirichlet boundary conditions ($u = v = 1$) were assumed, and it was found that periodic

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solutions appear when Le is small enough. The initial appearance of these oscillations as Le is lowered from 1 seems to result from a Hopf bifurcation, but as Le decreases further, the sinusoidal character of the solution is lost.

In [9] W. H. Ray and the present writer suggested that these periodic solutions tend to a "relaxation oscillation" as Le $\to$ 0. We studied a related model, the ordinary differential equations of a continuous stirred tank reactor (CSTR) in some detail, and noted qualitative similarities of this system to van der Pol's equation. (The scaling and notation in that paper were different from those used here.) A related study was done by Chang and Calo [4]. Both of these references concentrate on spatially homogeneous situations, though in [9] some conjectures are made concerning spatially inhomogeneous solutions. the purpose of this paper is to give a more complete analysis of a spatially non-uniform configuration.

The numerical integrations of Lee and Luss were done with $\gamma = 30$, which is "large" in this context. It was found that the solution changes slowly for all but about 6% of the cycle, while during this relatively short interval a "hot spot" develops near $x = 0$ and spreads rapidly toward $x = 1$, with very steep spatial gradients being exhibited for a brief time.

An appropriate averaging, or "lumping" technique can be used on Eqs. (1)–(5) to arrive at a pair of ordinary differential equations resembling those of the CSTR, and as in [8, 9], relaxation oscillations are found. Four distinct sub-intervals of a single period can be identified, two of length $O(\mathrm{Le})$ as Le $\to 0$ for fixed γ, one of length $O(1/\gamma)$ as $\gamma \to \infty$ uniformly in Le, and one of length $O(e^{-\gamma}/\gamma^2)$ as $\gamma \to \infty$, also uniformly in Le. These four intervals are not apparent in the computations of Lee and Luss, probably because numerical stiffness prevented choosing Le smaller than $1/10$.

We do not carry out a careful asymptotic analysis of the p.d.e.'s, though if feasible this would be a worthwhile project. The treatment here is too coarse to explain the hot spot. Instead, a somewhat simplified model is studied in enough detail to make the relaxation character of the oscillations clear. This is done formally, giving what amounts to the lowest order term of an expansion of the periodic solution in powers of Le. A simple graphical method is presented for estimating how large γ must be to support oscillations for given β and φ. A few computations are included to show that periodic solutions occur for reasonable values of β, γ, and φ, if Le is small enough. Estimates of the period and amplitude of the oscillation for large γ are presented. Well known results can be used to obtain a more accurate asymptotic estimate of the period as $\gamma \to \infty$.

II. Formal relaxation oscillations. A prototype for the study of relaxation oscillations is van der Pol's equation $\ddot{y} + \mu(y^2 - 1)\dot{y} + y = 0$. Setting $s = \varepsilon t$, $U(s) = -\varepsilon\int_0^t y(r)\,dr$, $V(s) = y(t)$, and $\varepsilon = 1/\mu$ leads to the equations

$$\begin{aligned} \varepsilon^2 V' &= V - (V^3/3) + U, \\ U' &= -V \end{aligned} \tag{6}$$

where "$'$" = "d/ds", and we assume (without loss of generality) that $\varepsilon y'(0) = y(0) - y(0)^3/3$. For $\varepsilon > 0$ this system has a unique orbitally asymptotically stable periodic

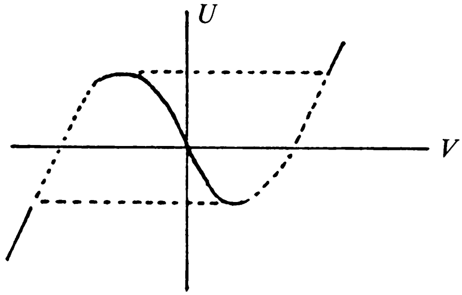

FIG. 1

solution, and as $\varepsilon \to 0$ the phase plane trajectory of this solution tends to the dotted curve in Fig. 1.

By a "formal relaxation oscillation" for (6) we mean a piecewise continuous curve $s \to (V(s), U(s))$ such that U and V are periodic with some smallest period $p > 0$ and the following conditions hold in $[0, p]$.

(a) $U = (V^3/3) - V$

(b) U is continuous, and V is continuous except at jump points s_1 and s_2 in $[0, p]$.

(c) $U'(s) = -V(s)$ except as s_1 and s_2.

(d) At s_1 and s_2, V jumps from a relative maximum or minimum of the cubic curve $U = (V^3/3) - V$ to a stable equilibrium point of the single equation

$$\varepsilon^2 V' = U(s_i) + V - V^3/3.$$

The precise form of the nonlinearity is not important, not is it necessary that the equation be linear. As a generalization of (6) consider a system

$$\begin{aligned} \varepsilon^2 V' &= U - f(V), \\ U' &= g(U, V). \end{aligned} \tag{7}$$

One could list properties of f and g which imply the existence of relaxation oscillations. In Fig. 2 we simply sketch a typical phase plane, including the curves $U = f(V)$, $g(U, V) = 0$, and a dotted curve which indicates the trajectory of a formal oscillation.

Observe that the equilibrium points of (7) are all on a decreasing segment of the curve $U = f(V)$. Otherwise the system may not support a relaxation oscillation because the equilibrium point could be stable.

Turning to (1)–(5), in order to discuss relaxation oscillations one must consider the "reduced system" (as Le $\to 0$), which consists of (1) together with a "steady state" problem in v, namely

$$\begin{gathered} v'' + \gamma\varphi^2 u F(v) = 0, \\ v'(0) = 0, \qquad v'(1) = \mu(1 - v(1)). \end{gathered} \tag{8}$$

This problem defines v as a function of x for each t, since $u(\cdot, t)$ varies with t according to (1).

Unfortunately it is difficult to analyze the solutions of (8) for various functions u, even if Dirichlet boundary conditions are assumed, and so we have been unable to prove the existence of a formal relaxation oscillation for (1)–(5).

III. A partial averaging. For this reason we now introduce a system consisting of an ordinary differential equation for the concentration u coupled with a partial differential equation for the temperature v. The model, which was apparently first given by N. R. Amundsen and L. R. Raymond [2], describes a situation in which "there is no resistance to mass transfer within the particle, and any resistance which may be present may be lumped at the surface... and characterized by a mass transfer coefficient. Heat generated within the particle must be conducted to the surface through the particle, the heat carried by the reactants and products being neglected." Upon rescaling it may also be viewed as an approximate model of a particle with very low temperature conductivity.

The equations are

$$\frac{du}{dt} = 1 - u - \varphi^2 u \int_0^1 F(v(x,t))\,dx, \quad (9)$$

$$\mathrm{Le}\frac{\partial v}{\partial t} = \frac{\partial^2 v}{\partial x^2} + \beta\varphi^2 u F(v), \tag{10}$$

$$\frac{\partial v}{\partial x}(0,t) = 0, \qquad v(1,t) = 1. \tag{11}$$

The simplified nondimensional form will suffice for a qualitative explanation of the oscillations. The Dirichlet condition on v at $x = 1$ results, formally, from letting $\mu \to \infty$ in (5). Although Amundsen and Raymond indicated that they found this system difficult to analyze, subsequent mathematical advances make (9)–(11) significantly easier to deal with than (1)–(5), at least in the limit as $\mathrm{Le} \to 0$. This will be seen further on.

The system (9)–(11) can be obtained from (1)–(5) by formal asymptotics. By rescaling in (1), a parameter $1/\eta$, $\eta \ll 1$, can be introduced in front of the diffusion term $\partial^2 u/\partial x^2$, while keeping Le small. It is also necessaru to let $\nu = \eta$ in (4), and take $v(1) = 1$ instead of (5). This approach is similar to that of D. Cohen and A. Poore in their analysis of a tubular reactor [5]. If a solution is sought in the form

$$u(x,t) = \sum_{i=0}^{\infty} u_i(x,t)\eta^i, \qquad v(x,t) = \sum_{i=0}^{\infty} v_i(x,t)\eta^i$$

then the lowest order terms u_0 and v_0 are found to satisfy (9)–(11). Unfortunately, it is not clear that the assumption $\eta \ll 1$ is physically justified in our case. Nevertheless, study of this system is worthwhile if it leads to greater understanding of the factors which can give periodicity in an exothermic chemical reactor. For example, our results suggests that oscillations can be driven by temperature gradients together with fluxes of reactant and heat at the boundary.

Mathematically the system (9)–(11) is still infinite dimensional, since it involves a partial differential equation, and we are not aware of previous demonstrations of formal relaxation oscillations for such a model.

Recent results of H. Matano [8] imply that if $(u(t), v(x, t))$ is a solution (9)–(11) and $v_x(x, 0) < 0$ on $(0, 1)$, then $v_x(x, t) \leqslant 0$ for all x in $[0, 1]$, $t > 0$. This implies that solutions of this system cannot exhibit the sort of hot spot seen by Lee and Luss, in which distinctly non-monotone temperature gradients are seen at times in the cycle.

IV. Formal relaxation oscillations for (9)–(11). By formally taking $\mathrm{Le} = 0$ in (1) we arrive at the boundary value problem

$$\begin{gathered} v'' + \varphi^2 uF(v) = 0, \\ v'(0) = 0, \qquad v(1) = 1. \end{gathered} \tag{12}$$

This is a much studied problem, with the most complete published results to date appearing in a recent article by N. Dancer [6]. He shows, for more general problems as well as (12), that solutions of (12) with $v(0) - 1 = O(1/\gamma)$ as $\gamma \to \infty$ are close to solutions of the so-called Gelfand problem

$$\begin{gathered} y'' + \delta e^{y} = 0, \\ y'(0) = y(1) = 0 \end{gathered} \tag{13}$$

where $y \sim \gamma(v - 1)$ for large γ. This problem can be solved exactly [3] and it is known that there is a critical value $\delta_c \approx .878$ such that (13) has two solutions if $\delta < \delta_c$ and no solutions if $\delta > \delta_c$. Dancer's work then gives complete asymptotic information about the "small" solutions of (12); that is, those solutions with $v(0) - 1 = O(1/\gamma)$. Recently J. B. McLeod [private communication] has obtained even more complete results, for all solutions of (12).

As a preliminary remark about (12), note that $F(v) > 0$, so that if v is a solution, then $v' < 0$ and $v'' < 0$ on $(0, 1)$.

Let $\Gamma = \{(u, v(\cdot)) \mid u \geqslant 0$ and (12) is satisfied$\}$. It is shown in [5] that Γ is a smooth curve in $R^+ \times C[0, 1]$ with end point $u = 0$, $v \equiv 1$. If $\hat{\Gamma}$ denotes the projection of this curve into R^2 given by the mapping $(u, v(\cdot)) \to (u, v(0))$, then it is easily seen that $\hat{\Gamma}$ is the graph of a function $u = \hat{u}(v(0))$. The definition of a formal relaxation oscillation can be given in an elementary manner using this function. (More sophisticated concepts could be used, but seem unnecessary here.) This definition parallels (a)–(d) above.

DEFINITION. *A formal relaxation oscillation for* (9)–(11) *is a piecewise continuous curve* $s \to (u(s), v(\cdot, s))$ *in* $R^+ \times C[0, 1]$ *with some period* $p > 0$ *such that the following conditions hold in* $[0, p]$.

(A) *The equations* (12) *hold for each* s. (In other words, $u(s) = \hat{u}(v(0, s))$.)

(B) *The function* u *is continuous, and* v *is continuous except at a finite number of jump points* $s_1, s_1, \ldots, s_m$ *in* $[0, p]$.

(C) *Equation* (9) *is satisfied except at the* s_i.

(D) *At each* s_i, $v(0, s)$ *jumps from a relative maximum or minimum of* $\hat{u}$ *to a point* v_0 *such that the solution of the initial value problem*

$$\begin{gathered} v'' + \beta\varphi^2 u(s_i)F(v) = 0 \\ v'(0) = 0, \qquad v(0) = v_0 \end{gathered} \tag{14}$$

is a stable steady state solution of the boundary problem

$$\mathrm{Le}\frac{\partial v}{\partial t} = \frac{\partial^2 v}{\partial x^2} + \beta\varphi^2 u(s_i)F(v), \qquad \frac{\partial v}{\partial x}(0,t) = 0, \qquad v(1,t) = 1. \tag{15}$$

(It is known that (15) has a least one stable steady state solution [1].)

In order to construct such a "singular solution" of (9)–(11) it is necessary to analyze $\hat{\Gamma}$, showing that it is "cubic like", and that the steady state solutions of the full system (9)–(11) lie, when projected, on a decreasing segment of $\hat{\Gamma}$. As suggested in the earlier discussion of ordinary differential equations (see Fig. 2), it is not necessary to show that $\hat{u}$ has exactly one maximum and one minimum. In an asymptotic sense, for sufficiently large γ, most of the necessary properties of $\hat{\Gamma}$ follow from known results about the Gelfand equation plus Dancer's work. However one key point, that steady states of the full system project onto a decreasing branch of $\hat{\Gamma}$, requires an extra computation. In fact, it is not difficult to obtain criteria which insure the existence of a formal relaxation oscillation for specific finite values of β, φ, and γ and which are easily checked numerically (even on a pocket calculator.) As a first step, integrate (12) twice to obtain

$$\hat{u}(v_0) = (v_0 - 1)\Big/\left(\beta\varphi^2\int_0^1 (1-s)F(v(s))\,ds\right)$$

with $v_0 = v(0)$. Since F is increasing and v is decreasing, it follows that

$$\hat{u}(v_0) \geqslant R(v_0) \equiv 2(v_0 - 1)\big/\beta\varphi^2 F(v_0). \tag{16}$$

On the other hand, since $v'' > 0$, we obtain $v(s) \geqslant v_0(1-s)$ and therefore

$$\hat{u}(v_0) \leqslant Q(v_0) \equiv (v_0 - 1)\Big/\left(\beta\varphi^2\int_0^1 ue^{\gamma(v_0-1)u/v_0}\,du\right). \tag{17}$$

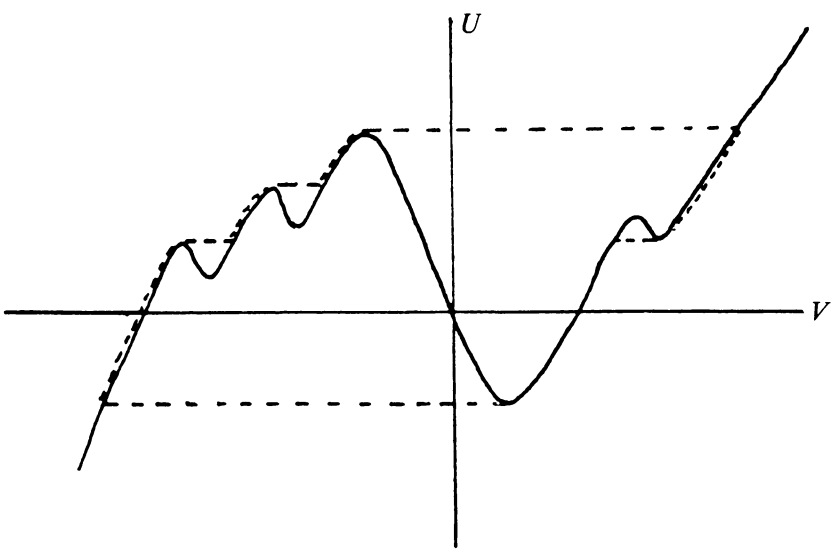

FIG. 2

It is easily computed that $R'(1)$ is positive and, if $\gamma > 4$, R' changes sign at

$$V_0^- = \frac{\gamma}{2}\left[1 - \sqrt{1 - 4/\gamma}\right] < \gamma/2$$

and at

$$V_0^+ = \frac{\gamma}{2}\left[1 + \sqrt{1 - 4/\gamma}\right] > \gamma/2.$$

Further, since $F(v_0) \to e^\gamma$ as $v_0 \to \infty$, R tends to infinity with v_0.

Further information on $\hat{\Gamma}$ is found by computing F' and F''. It is found that $F' > 0$, and $F''(v) > 0$ if $1 < v < \gamma/2$. It is known [1] that under these circumstances, for a given $u > 0$, (12) can have no more than two solutions with $1 \leqslant v(0) \leqslant \gamma/2$. Hence $\hat{u}$ can have at most one (relative) maximum, and no relative minima, in $(1, \gamma/2)$.

To establish that $\hat{u}$ has at least one maximum and at least one minimum, it is sufficient to check that

$$Q(v_1) < R(v_0^-) \tag{18}$$

for some $v_1 > v_0^-$.

Note that this condition does not depend on β or φ. It is easily computed numerically, after computing Q in closed form, and holds for γ greater than about 6.56. (See Fig. 3.)

The next step is to analyze steady states of the complete system (9)–(11). If $(u, v(\cdot))$ is such a steady state, then

$$\begin{aligned} u &= 1\Big/\left(1 + \varphi^2\int_0^1 F(v(s))\,ds\right) \\ &\geqslant P(v_0) \equiv 1/\left(1 + \varphi^2 F(v_0)\right), \qquad v = v(0). \end{aligned}$$

Since (v_0, u) also lies on $\hat{\Gamma}$, it must be to the right of the first intersection of the graphs of P and Q. On the other hand,

$$\begin{aligned} v_0 &= 1 + \beta\varphi^2 u\int_0^1 (1-s)F(v(s))\,ds \\ &= 1 + \left(\beta\varphi^2\int_0^1 (1-s)F(v(s))\,ds\Big/\left(1 + \varphi^2\int_0^1 F(v(s))\,ds\right)\right) \\ &\leqslant 1 + \beta. \end{aligned}$$

All of this leads to the following result.

Theorem 1. Suppose that $\gamma > 4$ and $1 + \beta < \gamma/2$. Let $v = v_*$ be the smallest solution of $P(v) = Q(v)$. If $v_* > v_0^-$ and $P(v_*) < R(v_0^-)$, then (9)–(11) admits a formal relaxation oscillation.

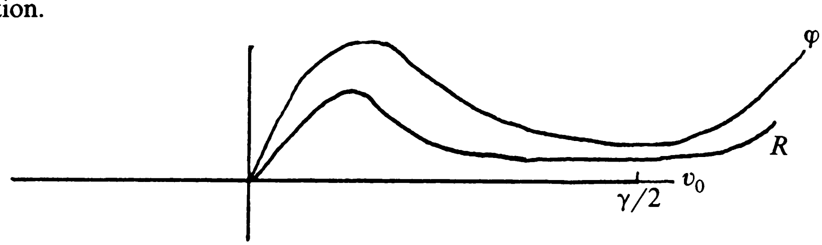

Fig. 3

Proof. Our previous remarks show that in $[1, \gamma/2)$, $\hat{u}$ has exactly one maximum, say at $v_0 = \hat{v}_0$, and no minima. Also, this function has at least one minimum in $[\gamma/2, \infty)$. Each (u, v_0) in $[0, \infty) \times [1, \infty)$ defines a unique solution $v(\cdot, u, v_0)$ of (13). The function

$$G(u, v_0) = 1 - u - \varphi^2 u \int_0^1 F(v(s, u, v_0))\, ds$$

is continuous, and indeed smooth, in (u, v_0). Consider the pair of equations

$$\begin{aligned} \varepsilon^2 V' &= U - \hat{u}(V), \\ U' &= G(U, V). \end{aligned} \tag{17}$$

Our hypotheses guarantee that all equilibrium points of this system lie in the region $1 < V < \gamma/2$ and to the right of $V = \hat{v}_0$. Therefore these solutions all lie on a single decreasing branch of $\hat{\Gamma}$. Further, $G(\hat{u}(V), V) > 0$ if $V \leqslant \hat{v}_0$ and $G(\hat{u}(V), V) < 0$ if $V \geqslant \gamma/2$. Hence a formal relaxation oscillation can be constructed for (16), and this corresponds in an obvious way to a formal relaxation oscillation for (9)–(11). This proves Theorem 1.

It is, of course, necessary to check that the hypotheses of Theorem 1 can be satisfied, and to say as much as possible about the values of β, φ and γ which give oscillations. A brief summary of some typical numerical results, covering some ranges of β and φ^2 which are given as feasible in [3], appears in Table 1. Values of γ up to around 30 have been reported in the literature.

TABLE 1. Smallest integer γ such that β, φ, γ satisfy hypotheses of Theorem 1.

	.1	.3	.6	.9
.5	130	38	24	19
1	130	38	24	16
4	130	38	19	16
7	130	38	19	16

It will be noted that the parameters chosen by Lee and Luss ($\beta = .15$, $\varphi = 1.1$, $\gamma = 20$) do not fall in the range where we predict oscillations. This may be because our estimates are conservative, or because of differences between their system and (9)–(11).

Theoretically we have the following result.

THEOREM 2. For any given $\beta > 0$ and $\varphi > 0$, the system (9)–(11) satisfies the hypotheses of Theorem 1 if γ is sufficiently large.

Proof. The lower root of R' is

$$v_0^- = 1 + 1/\gamma + o(1/\gamma) \quad \text{as } \gamma \to \infty,$$

and

$$R(v_0^-) = (2/\beta\varphi^2 e\gamma) + o(1/\gamma). \tag{19}$$

Also

$$F(v_0^-) = e + o(1)$$

Hence on $[1, v_0^-]$, $P(v_0) \geq 1/(1 + \varphi^2 e) + o(1)$.

On the other hand, it is easily shown that if $1 \leq v_0 \leq v_0^-$, then

$$\varphi(v_0) \leq (2/\beta\varphi^2\gamma) + o(1/\gamma).$$

Hence on this interval $Q(v_0) < P(v_0)$ if γ is sufficiently large.

LEMMA 1. At an equilibrium point $(u, v(\cdot))$ of (9)–(11), $(Q(\gamma/2)) < u < R(v_0^-)$ if γ is sufficiently large.

Proof. The first inequality holds because $v_0 < 1 + \beta$ and $Q(\gamma/2) = O(e^{-\gamma}/\gamma^2)$, while $R(1 + \beta) = O(1/e^{\gamma(\beta/(1+\beta))})$. For the second, consider a v_0 such that $P(v_0) = Q(v_0)$. Let $\alpha = \gamma(v_0 - 1)/v_0$. Integrating to find Q in closed form shows that

$$1/\left(1 + \varphi^2 F(v_0)\right) = \alpha^3 v_0/\varphi^2\gamma\left[(\alpha - 1)e^{\alpha} + 1\right].$$

If $v_0 \geq v_0^-$ then $\alpha \geq 1 + o(1/\gamma)$ so that either $\alpha \geq 1$, or $|1 - \alpha| = o(1/\gamma)$ and $e^{\alpha} \leq e$. In either case, since $v_0 \leq 1 + \beta$, we find that

$$\alpha^3 \geq \hat{K}\gamma$$

for some $\hat{K}$ which is independent of γ. This leads to $v_0 \geq 1 + (K/\gamma^{2/3})$ for some positive K which is independent of γ. To complete the proof of Lemma 1 and Theorem 2 we show that $Q(v_0) < R(v_0^-)$ if $1 + (K/\gamma^{2/3}) \leq v_0 \leq 1 + \gamma$ and γ is sufficiently large. First identify an interval, which includes $[1 + (K/\gamma^{2/3}), 1 + \beta]$, on which Q is decreasing.

LEMMA 2. For large enough γ, Q is decreasing on

$$\left[1 + 3(1 + \beta)^2/2\gamma, 1 + \beta\right].$$

Proof. It is easily computed that $Q'(v_0) < 0$ if

$$(v_0 - 1)/v_0 > \int_0^1 ue^{\alpha u}\,du \Big/ \gamma\int_0^1 u^2 e^{\alpha u}\,du, \qquad \alpha = \gamma(v_0 - 1)/v_0,$$

and further that $\int_0^1 ue^{\alpha u}\,du/\int_0^1 u^2 e^{\alpha u}\,du \leq 3/2$ for any $\alpha \geq 0$. The result follows.

Hence we must show that $Q(1 + K/\gamma^{2/3}) < R(v_0^-)$ for large γ. But

$$Q(1 + K/\gamma^{2/3}) = K\Big/\left[\gamma^{2/3}\beta\varphi^2\int_0^1 ueK\gamma^{+1/3}u/\left(1 + K\gamma^{-2/3}\right)du\right]$$

$$= o(1/\gamma) \quad \text{as } \gamma \to \infty.$$

Using (19), this completes the proofs of Lemma 1 and Theorem 2.

V. Asymptotic estimates of the period and amplitude. It is not difficult to obtain asymptotic estimates, as $\gamma \to \infty$, for the period and amplitude of the formal relaxation oscillation found above. It is first necessary to estimate $Q_{\max} = \max_{1 \leq v_0 \leq 1+\beta} Q(v_0)$.

It was shown in Lemma 2 that this maximum is taken on in the interval $1 \leq v_0 \leq 1 + M/\gamma$ where $M = 3(1 + \beta)^2/2$. Setting $Q'(v_0) = 0$ gives

$$\int_0^1 \left(u - \left(\gamma(v_0 - 1)u^2/v_0\right)\right)e^{\gamma(v_0-1)u/v_0}\,du = 0$$

from which

$$Q(v_0) = v_0 \Big/ \left(\beta\varphi^2\gamma\int_0^1 u^2 e^{\gamma(v_0-1)u/v_0}\right) du$$
$$\leqslant (3/\beta\varphi^2) + O(1/\gamma). \tag{20}$$

Let $s \to (u(s), v(\cdot, s))$ be the relaxation oscillation. We can assume that $u' > 0$ on an interval $(0, T_1)$ and $u' < 0$ on (T_1, p), where p is the period. The amplitude of the oscillation in u is $A = u(T_1) - u(0)$. Figure 4 shows a possible projection of this "singular solution" onto the $(u, v(0))$ plane.

It is clear that $R(v_0^-) \leqslant u(T_1) \leqslant Q_{\max}$ and $0 < u(0) \leqslant Q(\gamma/2)$. Our estimates on Q and R then show that

$$\frac{2}{\beta\varphi^2 e\gamma} + o\left(\frac{1}{\gamma}\right) \leqslant A \leqslant \frac{3}{\beta\varphi^2\gamma} + o\left(\frac{1}{\gamma}\right)$$

as $\gamma \to \infty$. The maximum value $v_{\max}$ of the non-dimensional temperature variable v lies between v_1 and $_2$, where these are defined by the relations $v_2 > v_1 > \gamma/2$, $Q(v_1) = R(v_0^-)$ and $R(v_2) = Q_{\max}$. To estimate v_1 we have

$$R(v_0^-) = (2/\beta\varphi^2 e\gamma) + o(1/\gamma) = (v_1 - 1)\Big/ (\beta\varphi^2)\int_0^1 u e^{\gamma(v_1-1)u/v_1} du$$
$$\leqslant (v_1 - 1)\Big/ \left(\beta\varphi^2 \int_0^1 u e^{(\gamma-2)u} du\right)$$

since $v_1 \geqslant \gamma/2$. This gives

$$v_1 \geqslant 2e^{\gamma}/(\gamma^2 e^3) + o(e^{\gamma}/\gamma^2). \tag{21}$$

Also,

$$Q_{\max} = \left[\frac{3}{\beta\varphi^2\gamma} + o\left(\frac{1}{\gamma}\right)\right] = 2(v_2 - 1)\Big/ e^{\gamma(v_2-1)/v_2} \geqslant 2(v_2 - 1)\Big/ e^{\gamma}$$

from which

$$v_2 \leqslant \frac{3}{2\beta\varphi^2}\frac{e^{\gamma}}{\gamma} + o(e^{\gamma}/\gamma). \tag{22}$$

The estimates (21) and (22) are somewhat unsatisfying, as it would be nice to pin down more precisely the order of growth of the maximum temperature with γ.

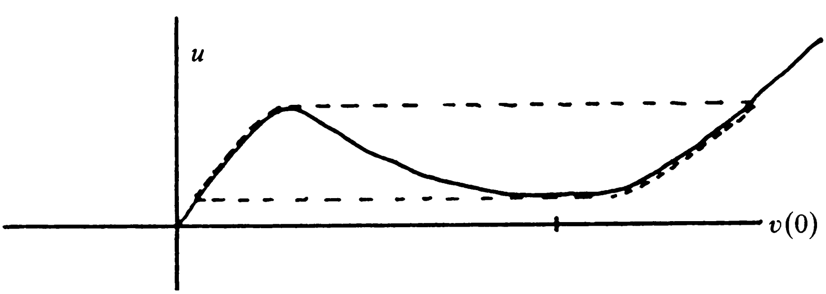

FIG. 4

Estimates of the period are also easily obtained from these inequalities, by showing that $u' = 1 + O(1/\gamma)$ on $(0, T_1)$ and $u' = -0(\gamma)$ on (T_1, p). This leads to the conclusion that $T_1 = p + O(1)$ and

$$\frac{2}{\beta\varphi^2 e\gamma} + O\left(\frac{1}{\gamma}\right) \leqslant p \leqslant \frac{3}{\beta\varphi^2\gamma} + o\left(\frac{1}{\gamma}\right).$$

A more accurate estimate of p is found from the exact solution of Gelfand's equation [3], showing that $p \sim .868/\beta\varphi^2\gamma$ as $\gamma \to \infty$. This gives, as well, a refined estimate of the amplitude A, but does not help improve on (21) and (22).

Finally, these estimates can be compared with the computations of Lee and Luss. For their values of the parameters the best estimate of period above gives $p \approx .16$, whereas their computed period is about $p = .25$. However the maximum amplitude in temperature found by Lee and Luss is many order of magnitudes lower than our estimate. This is almost certainly due to the extreme stiffness of the system for small Lewis number. They were unable to carry the computations below Le $= .1$, and indeed, it is not clear that very much more could be achieved today.

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