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1976 J. Phys. A: Math. Gen. 9 77

(http://iopscience.iop.org/0305-4470/9/1/013)

J. Phys. A: Math. Gen., Vol. 9, No. 1, 1976. Printed in Great Britain. © 1976

On the spectral theory of Rayleigh's piston III. Exact solution of the absorbing barrier problem ($\gamma = 1$)

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Received 28 April 1975, in final form 9 September 1975

Abstract. We derive an exact solution for the evolution of a one-dimensional ensemble of Rayleigh test particles at unit mass ratio ($\gamma = 1$) in the presence of a *speed* absorbing barrier. The result, which entails an Arrhenius-law dependence of mean first-passage time on barrier height, appears to be a hitherto unique example of a singular passage-time problem exactly soluble through a continuous spectrum of eigenvalues and their associated eigen-distributions.

1. Introduction

In our earlier papers in this series (Hoare and Rahman 1973, 1974, to be referred to as I and II respectively) we developed a qualitative treatment of the spectral properties of the Rayleigh one-dimensional gas and obtained an exact solution of the singular eigenvalue problem for the relaxation modes in the special case of unit test particle/heat bath mass ratio. This led to a simple closed form for the evolution of the *speed* distribution function, though the behaviour of the more interesting *velocity* distribution remained very complicated.

We now turn to an aspect of the Rayleigh problem which, so far as we are aware, has never been considered either in this model or in any comparable formulation of particle transport theory. This concerns the evolution of a system of Rayleigh test particles in the presence of an *absorbing barrier* representing the physical removal of those particles which reach some critical threshold in speed or energy. Such a situation seems not without practical importance—one can envisage both experimental and astrophysical conditions under which particles might be removed to some *null state* by chemical, or conceivably nuclear, reaction at a certain activation barrier and thereafter play no further part in the process. The instantaneous initiation of the process is less easily allowed for, but might nevertheless be achieved by sudden irradiation or exposure to a shock front.

A considerable literature deals with this type of system in the much simpler case, appropriate only to *internal* degrees of freedom, where the interaction with a heat bath is specified in terms of a mean collision number and the reaction process can be considered in terms of discrete 'relaxation times' (Montroll and Shuler 1958, Widom 1959, Hoare 1964, Oppenheim *et al* 1967). In these studies particular attention is paid to the dependence of overall rate of reaction (absorption) upon both barrier energy and the initial distribution of reactant particles, and it is a notable feature of all models so far treated that the existence of a characteristic first-order rate constant with exponential dependence on barrier height (the Arrhenius law) can only be justified asymptotically for limitingly high barrier energy. When, as here, reaction is required to depend on the *translational* degree of freedom, the occurrence of singular solutions and a continuous distribution of relaxation times rules out the simple treatment previously applied, though similar questions to those above remain of interest in the resulting, more complicated, mathematical picture.

The present work appears to be the first to examine these problems—in particular the status of the Arrhenius law—in relation to a singular kinetic process with absorption. While the one-dimensional Rayleigh model can hardly be expected to provide direct predictions relevant to actual reactions, for example in a three-dimensional shock front, it may nevertheless be offered as an idealized case of exceptional interest which can throw light on the mathematics of more realistic systems.

2. The transport equation

For physical and mathematical background to the Rayleigh piston model we refer to I and II. Once again we restrict attention to the special case ($\gamma = 1$) in which labelled test particles collide with a one-dimensional heat bath of particles of identical mass, and consider the half-range initial-value problem for the evolution of a *speed* distribution function $P(|x|, \tau)$ given a specified $P(|x|, 0)$. Let there now be introduced an *absorbing barrier* at some speed $x^\dagger$ such that particles once exceeding this 'react' and are removed from the system. Using the transition probabilities treated earlier (equations (5.5) and (5.6) of II), the transport equation to be satisfied by a time-dependent distribution $P(|x|, \tau)$ is easily seen to be†

$$\frac{\partial}{\partial \tau} P(|x|, \tau) = 2\,\mathrm{e}^{-x^2} \int_0^{x^\dagger} \max(x, y) P(y, \tau)\, \mathrm{d}y - z(x) P(|x|, \tau) \qquad (0 \leqslant x \leqslant x^\dagger) \qquad (2.1)$$

in which the collision number function $z(x)$ is

$$z(x) = \mathrm{e}^{-x^2} + \pi^{1/2} x\, \mathrm{erf}(x). \qquad (2.2)$$

Here $\max(x, y)$ represents the value of the larger of the two arguments. We recall that

$$z(0) = 1, \qquad z'(x) = \pi^{1/2}\, \mathrm{erf}(x), \qquad z''(x) = 2\,\mathrm{e}^{-x^2}.$$

As we showed previously (II, equation (9.1)) the 'fundamental' solution of the unperturbed relaxation problem ($x^\dagger = \infty$) corresponding to the initial condition $P(x, 0) = \delta(x - x_0)$ can be written in the form

$$P(x, \tau) = 2\pi^{-1/2}\,\mathrm{e}^{-x^2} + \delta(x - x_0)\,\mathrm{e}^{-z(x_0)\tau} - 2\tau\,\mathrm{e}^{-x^2} \int_{\max(x, x_0)}^{\infty} \mathrm{e}^{-z(y)\tau}\, \mathrm{d}y; \qquad \tau > 0. \qquad (2.3)$$

In fact the general solution for arbitrary normalized initial condition $P(x, 0)$ can be obtained by a similar method, or equally well by superposition of the solutions (2.3).

† Throughout this paper x is the reduced velocity $x = (M/2k_{\mathrm{B}}T)^{1/2} V$, where M, T, V are the common test particle and heat bath mass, heat bath temperature and test particle velocity respectively. $\tau = n\sigma(2k_{\mathrm{B}}T/\pi M)^{1/2} t$ with n, σ test particle number density and collision cross section. We shall drop the modulus notation from $|x|$ in what follows on the understanding that all expressions refer strictly to the half-range $0 \leqslant x \leqslant x^\dagger$ with $x^\dagger < \infty$.

This reads

$$P(x,\tau)=2\pi^{-1/2}\,\mathrm{e}^{-x^2}+P(x,0)\,\mathrm{e}^{-z(x)\tau}-2\tau\,\mathrm{e}^{-x^2}\int_x^{\infty}\mathrm{e}^{-z(y)\tau}\int_0^y P(w,0)\,\mathrm{d}w\,\mathrm{d}y$$

$$\tau>0. \tag{2.4}$$

Our objective in this paper is to derive a similar expression for the perturbed distribution $P(x, x^\dagger, \tau)$ in the presence of an absorbing barrier at speed $x^\dagger$. This must evidently express the loss of conservation of probability over the finite range $(0, x^\dagger)$ through a modification of the persistent, Maxwellian term in the above equations.

3. The eigenvalue problem

Following our previous development of the solution of (2.4) we may expect the perturbed distribution function $P(x, x^\dagger, \tau)$ solving equation (2.1) to be expressible as a combination of terms of the form

$$P(x, x^\dagger, \tau)\sim\exp[-\lambda(x^\dagger)\tau-x^2]f(x, x^\dagger, \lambda) \tag{3.1}$$

where $f(x, x^\dagger, \lambda)$ is to be determined by solution of the singular integral equation

$$[z(x)-\lambda]f(x, x^\dagger, \lambda)=2\int_0^{x^\dagger}\max(x, y)f(y, x^\dagger, \lambda)\,\mathrm{d}y. \tag{3.2}$$

Although in outline the solution of this may be expected to parallel that for the simple relaxation problem, to which it reduces for $x^\dagger=\infty$, the presence of the absorbing barrier truncating the kernel introduces new boundary conditions which must perturb the whole eigenvalue spectrum. There are two distinct consequences. First the equilibrium eigenvalue $\lambda_0=0$ of the simple relaxation operator will be perturbed to a finite value, thus removing the persistent term in the solution; secondly the restriction of the argument to the interval $0\leqslant x<x^\dagger$ reduces the previously infinite range of the continuous spectrum to the finite region $1<\lambda<z(x^\dagger)$ for which the singular factor $[z(x)-\lambda]$ can vanish (figure 1). We must also allow for the fact that the presence of the barrier might induce new discrete lines in the previously empty discretum region $0<\lambda<1$.

As before we first examine the possibility of solutions to (3.2) in the region of discrete eigenvalues $0<\lambda<1$. The general form of these is evidently unchanged by the condition $x^\dagger\neq\infty$ since we may differentiate twice and obtain the singular differential equation

$$(\mathrm{d}^2/\mathrm{d}x^2)\{[z(x)-\lambda]f(x, x^\dagger, \lambda)\}=2\mathrm{e}^{-x^2}f(x, x^\dagger, \lambda) \tag{3.3}$$

essentially as before. We note likewise that the condition $z''(x)=2\mathrm{e}^{-x^2}$ ensures that one solution of the above shall be of the form $f_0=$ constant, with the corresponding eigenvalue λ_0 to be fixed in terms of $x^\dagger$ through equation (3.2). A second solution may then be determined as

$$R(x,\lambda)=\int_0^x\frac{\mathrm{d}y}{[z(y)-\lambda]^2} \tag{3.4}$$

so that the general solution will take the form of a linear combination

$$f(x, x^\dagger, \lambda)=a(x^\dagger, \lambda)+b(x^\dagger, \lambda)\int_0^x\frac{\mathrm{d}y}{[z(y)-\lambda]^2}, \tag{3.5}$$

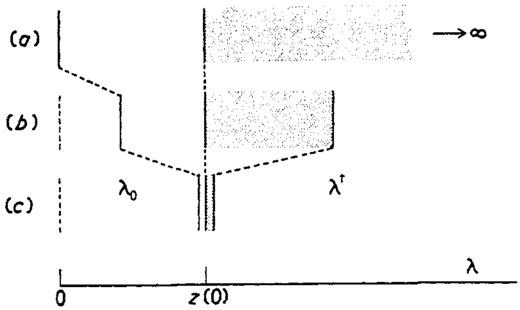

Figure 1. Eigenvalue spectrum related to barrier height for the Rayleigh scattering operator truncated at an absorbing barrier ($\gamma = 1$). (a) Spectrum for unperturbed relaxation ($x^\dagger = \infty$); (b) intermediate case; (c) spectrum for limiting region $x^\dagger \ll 1$. Note how the single discrete eigenvalue λ_0 is perturbed from its zero value by the introduction of the barrier, while the continuum is reduced to a finite range bounded above at $\lambda^\dagger = z(x^\dagger)$. The continuum threshold remains at $z(0)$ and thus the whole continuum and the single discrete line coalesce at the point $\lambda = z(0)$ as $x^\dagger \to 0$ (schematic only).

with two constants dependent on the barrier height $x^\dagger$. The integral is of course regular since we have restricted attention to the region $0 \leqslant \lambda < 1$. One of the constants will later be fixed by a normalization; to determine the second we substitute (3.5) back into the original integral equation (3.2) and obtain, after some partial integrations, the condition

$$-a(x^\dagger, \lambda) = a(x^\dagger, \lambda)[x^\dagger z'(x^\dagger) - z(x^\dagger)] + b(x^\dagger, \lambda)\Big[x^\dagger z'(x^\dagger) R(x^\dagger, \lambda) - z(x^\dagger) R(x, \lambda) + \lambda R(x^\dagger, \lambda) + \frac{x^\dagger}{z(x^\dagger) - \lambda} - \frac{x}{1-\lambda}\Big]. \tag{3.6}$$

Inspection shows that this can only be satisfied by forcing

$$b = 0; \qquad \lambda_0(x^\dagger) = z(x^\dagger) - x^\dagger z'(x^\dagger) \tag{3.7}$$

with the constant a remaining arbitrary. Using now the derivative $z'(x) = \pi^{1/2}\,\mathrm{erf}(x)$ from (2.2) the single discrete eigenvalue is revealed to be

$$\lambda_0(x^\dagger) = \mathrm{e}^{-x^{\dagger 2}}. \tag{3.8}$$

The rest of the discretum must be empty as in the unperturbed relaxation, confirming that no new eigenvalues are induced by the absorbing barrier.

The continuum region $\lambda > 1$ may now be considered. Following our previous solution for the unperturbed relaxation (II, equation (4.4)) we may expect the singular 'eigendistributions' of the operator in equation (3.2) to be expressible in the form

$$f(x, x^\dagger, \lambda) = \begin{cases} A_1\,\delta[z(x) - \lambda] + B_1 + C_1 R(x, \lambda); & 0 < x_\lambda \leqslant x < x^\dagger \\ A_2 + C_2 R(x, \lambda); & 0 \leqslant x < x_\lambda < x^\dagger \end{cases} \tag{3.9}$$

where R now stands for the pseudofunction Pf . $R(x, \lambda)$ corresponding to the integral of equation (3.4), x_λ is the root of $z(x_\lambda) - \lambda = 0$, and the constants appearing are all functions of λ. However, knowing that the present half-range problem requires solutions of *even* symmetry, we may reject the terms in $R(x, \lambda)$ and by substitution back into (3.2) establish in addition that $B_1 = 0$. At the same time the constants A_1 and A_2 prove to be related by the condition

$$A_1(\lambda) = -A_2(\lambda)\, e^{-x_\lambda^2}/z'(x_\lambda)^2. \tag{3.10}$$

Thus the required singular solutions (eigendistributions) can be written in terms of the one remaining normalization function $A(\lambda)$

$$f(x, x^\dagger, \lambda) = A(\lambda) \times \begin{cases} \delta[z(x)-\lambda]; & 0 < x_\lambda \leq x < x^\dagger \\ -\dfrac{e^{-x_\lambda^2}}{z'(x_\lambda)^2}; & 0 < x \leq x_\lambda < x^\dagger. \end{cases} \tag{3.11}$$

While the quantity $x^\dagger$ does not appear explicitly in the functions on the right we retain it as an argument to emphasize that it imposes a domain $(0, x^\dagger)$ on the new functions $f(x, \lambda)$ in contrast to the domain $(0, \infty)$ in the simple relaxation case.

The orthogonality of the above functions to the Maxwellian is readily confirmed, as is the orthogonality with respect to each other:

$$\int_0^{x^\dagger} e^{-x^2} f(x, x^\dagger, \lambda) f(x, x^\dagger, \lambda')\, dx = \frac{A(\lambda)^2\, e^{-x_\lambda^2}}{2z'(x_\lambda)}\, \delta(x - x_\lambda). \tag{3.12}$$

Thus we may work with the orthonormal set

$$\phi(x, x^\dagger, \lambda) = e^{-\frac{1}{2}x^2}\sqrt{2} \times \begin{cases} e^{x_\lambda^2} z'(x_\lambda)^2\, \delta[z(x)-\lambda]; & 0 < x_\lambda \leq x^\dagger \\ -1; & 0 < x \leq x_\lambda < x^\dagger. \end{cases} \tag{3.13}$$

The conditions on the right are, of course, equivalent to the restriction $1 \leq \lambda < z(x^\dagger)$.

We can now discern the full nature of the spectrum in the presence of the absorbing barrier. Since, as $x^\dagger \to 0$, $\lambda_0(x^\dagger) \to z(0) = 1$, we see that the single discrete line approaches the continuum threshold as the barrier height $x^\dagger$ is reduced, while at the same time the continuum itself is compressed into a narrowing region which eventually coalesces with the discrete line at $\lambda = 1$ (figure 1). This corresponds to the unrealistic limiting case where only those test particles of virtually zero velocity are considered and these are removed with a single time constant equal to their inverse collision number.

4. The initial-value problem

Usually the solution of stochastic problems with absorption is considerably more difficult than in the case of simple relaxation and we may have to be content with lesser information than the full time-dependent distribution function. Weaker alternatives are to seek the *passage-time distribution* for absorption, or its *moment generating function*, or in the simplest resort, the *mean first-passage time* to the absorbing barrier. Nevertheless, in our present case, in which the absorbing threshold $x^\dagger$ does not appear explicitly in the eigenfunctions, it proves possible to obtain the full distribution function $P(x, x^\dagger, \tau)$ itself as well as the reduced quantities.

To derive this we write a spectral expansion in the form

$$P(x, x^\dagger, \tau) = \alpha_0\, e^{-x^2 - \lambda_0(x^\dagger)\tau} + e^{-\frac{1}{2}x^2} \int_1^{z(x^\dagger)} \alpha(\lambda)\phi(x, \lambda)\, e^{-\lambda\tau}\, d\lambda \tag{4.1}$$

and, entering the eigenfunctions (3.13) obtain, for $\tau = 0$

$$P(x, x^{\dagger}, 0) = \alpha_0\,\mathrm{e}^{-x^2} + \alpha(x) z'(x)^2 - 2\,\mathrm{e}^{-x^2} \int_x^{x^{\dagger}} \alpha(x_\lambda) z'(x_\lambda) \phi(x, x_\lambda)\,\mathrm{d}x_\lambda. \tag{4.2}$$

Differentiation with respect to x leads to a first-order differential equation for the function $\alpha(x)$, for which the integrating factor is seen to be $z'(x)\exp(x^2)$, and this in turn leads to the solution

$$\alpha(x) = \frac{P(x, x^{\dagger}, 0)}{z'(x)^2} - \frac{2\,\mathrm{e}^{-x^2}}{z'(x)^3} \int_0^x P(y, x^{\dagger}, 0)\,\mathrm{d}y + \frac{\beta\,\mathrm{e}^{-x^2}}{z'(x)^3} \tag{4.3}$$

with β a constant of integration. Substitution back into equation (4.3) exposes the connection

$$\beta = 2 \int_0^{x^{\dagger}} [P(x, x^{\dagger}, 0) - \alpha_0\,\mathrm{e}^{-x^2}]\,\mathrm{d}x. \tag{4.4}$$

Two conditions now suffice to determine β and α_0. We impose the normalization

$$\int_0^{x^{\dagger}} P(x, x^{\dagger}, 0)\,\mathrm{d}x = 1 \tag{4.5}$$

and require that $\alpha(\lambda)$ is identically zero when $P(x, x^{\dagger}, 0)$ is the Maxwellian normalized on $(0, x^{\dagger})$. This leads to the combination

$$\beta = 0; \qquad \alpha_0 = 2/z'(x^{\dagger}). \tag{4.6}$$

We note that equation (4.3) amounts to a constructive proof of the completeness of the set (3.13) with respect to all functions simply integrable on $(0, x^{\dagger})$.

The solution of the initial-value problem for any probability distribution $P(x, x^{\dagger}, 0)$ may now be composed. Substituting the expression (4.3) for $\alpha(\lambda)$ back into (4.1) we obtain, after some manipulation

$$P(x, x^{\dagger}, \tau) = \frac{2\,\mathrm{e}^{-x^2}}{z'(x^{\dagger})}[\exp(-\tau\,\mathrm{e}^{-x^{\dagger 2}}) - \mathrm{e}^{-z(x^{\dagger})\tau}] + P(x, x^{\dagger}, 0)\,\mathrm{e}^{-z(x)\tau}$$
$$- 2\tau\,\mathrm{e}^{-x^2} \int_x^{x^{\dagger}} \mathrm{e}^{-z(y)\tau} \int_0^y P(w, x^{\dagger}, 0)\,\mathrm{d}w\,\mathrm{d}y. \tag{4.7}$$

Results for a variety of special cases follow immediately from this expression. Using, as necessary, the limiting values of the collision-number function:

$$z(0) = 1; \qquad z(\infty) = \infty; \qquad z'(0) = 0; \qquad z'(\infty) = \pi^{1/2}$$

we notice the following.

(i) For any initial condition normalizable by (4.5) we have

$$P(x, x^{\dagger}, \tau) \xrightarrow[\tau \to 0]{} P(x, x^{\dagger}, 0)$$

explicitly.

(ii) For long times

$$P(x, x^{\dagger}, \tau) \xrightarrow[\tau \to \infty]{} 0; \qquad x < x^{\dagger} < \infty.$$

(iii) For limiting low barrier height $x^\dagger \ll 1$, absorption occurs at first collision and

$$P(x, x^\dagger, \tau) \simeq P(x, x^\dagger, 0)\, e^{-z(x)\tau}.$$

(iv) For limiting high barrier $x^\dagger \to \infty$ the simple relaxation solution (2.4) is recovered.

(v) For the 'fundamental' initial condition $P(x, x^\dagger, 0) = \delta(x - x_0)$, the solution reduces to

$$P(x, x^\dagger, \tau) = \frac{2\,e^{-x^2}}{z'(x^\dagger)}[\exp(-\tau\, e^{-x^{\dagger 2}}) - e^{-z(x^\dagger)\tau}] + \delta(x - x_0)\, e^{-z(x_0)\tau}$$

$$-2\tau\, e^{-x^2} \int_{\max(x,x_0)}^{x^\dagger} e^{-z(y)\tau}\, dy \qquad (4.8)$$

which in turn gives the result (2.3) when $x^\dagger \to \infty$.

(vi) For an initial Maxwellian distribution at the heat-bath temperature, ie when the absorbing barrier is instantaneously 'switched on', there is cancellation of terms with the simple result that

$$P(x, x^\dagger, \tau) = \frac{2\,e^{-x^2}}{z'(x^\dagger)} \exp(-\tau e^{-x^{\dagger 2}}). \qquad (4.9)$$

Results computed from equation (4.8) with a number of different initial conditions and barrier heights are illustrated in figure 2.

The general content of the exact solution (4.7) is readily perceived. Evidently the subset of test particles which have experienced no collision up to time τ decays as a Poisson process with time constant $z(x)^{-1}$, this being the mean waiting time to collision at speed x. A complicated transient behaviour accompanies this with test particles taking up the Maxwellian distribution in competition with passage over the barrier. If the barrier height is very low, the integral in (4.7) makes little contribution; at the other extreme, when $x^\dagger \gg 1$, the system behaves at first as though undergoing simple relaxation, except that the resulting Maxwellian decays with a very long time constant $\exp(x^{\dagger 2})$ after all other transients have died out. We may note that this behaviour corresponds to the *Arrhenius law* of reaction kinetics, the quantity $x^{\dagger 2}$ representing a reduced *activation energy* and reaction proceeding in accordance with the *equilibrium hypothesis* (see eg Montroll and Shuler 1958). It is a notable feature of the Rayleigh model, however, that the Arrhenius law holds throughout *whatever* the form of the initial condition $P(x, x^\dagger, 0)$. Thus, on integrating equation (4.7) over all $x < x^\dagger$ to determine the total proportion $C(\tau)$ of particles unreacted by time τ, we find a cancellation similar to that in case (vi) above with the result

$$C(\tau) = \int_0^{x^\dagger} P(x, x^\dagger, \tau)\, dx = \exp(-\tau\, e^{-x^{\dagger 2}}). \qquad (4.10)$$

This implies equally simple behaviour for the distribution of first-passage times to absorption and its mean. Let these quantities be $w_1(\tau)$ and $\langle \tau_1 \rangle$ respectively. Then we have

$$w_1(\tau) = -dC(\tau)/d\tau = e^{x^{\dagger 2}} \exp(-\tau\, e^{-x^{\dagger 2}}) \qquad (4.11)$$

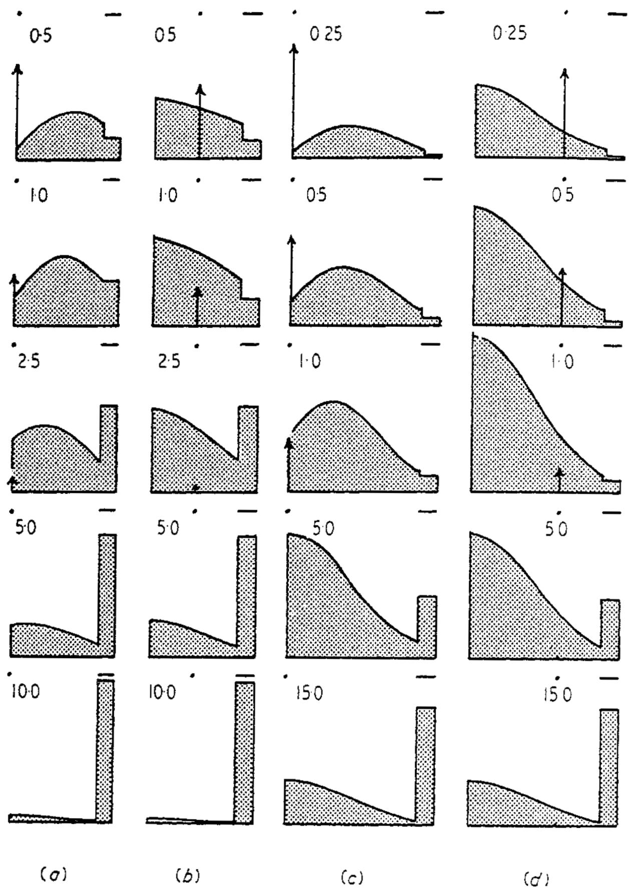

Figure 2. Relaxation of various delta ensembles of Rayleigh test particles in the presence of an absorbing barrier (equation (4.8)). Positions of initial delta function, x_0, and absorbing barrier, $x^{\dagger}$, in the four cases are: (*a*) $x_0 = 0{\cdot}0$, $x^{\dagger} = 1{\cdot}0$; (*b*) $x_0 = 0{\cdot}5$, $x^{\dagger} = 1{\cdot}0$; (*c*) $x_0 = 0{\cdot}0$, $x^{\dagger} = 1{\cdot}5$; (*d*) $x_0 = 1{\cdot}0$, $x^{\dagger} = 1{\cdot}5$. The vertical arrows represent the decay of the delta function, its probability component scaled to unity by the dot. The column on the right represents the integrated flux over the barrier, scaled to unity by the horizontal bar. The figures give the elapsed time in reduced units. Note the interplay between three effective time scales involving: (i) the decay of the delta function; (ii) relaxation to the Gaussian; (iii) leakage across the barrier. The distributions of unabsorbed test particles are effectively Gaussian for $\tau \gtrsim 5{\cdot}0$ in cases (*a*) to (*c*) and $\tau \gtrsim 1{\cdot}0$ in (*d*).

whence

$$\langle \tau_1 \rangle = \int_0^\infty \tau w_1(\tau)\, \mathrm{d}\tau = \int_0^\infty C(\tau)\, \mathrm{d}\tau = \mathrm{e}^{x^{\dagger 2}} \; (=\lambda_0^{-1}). \tag{4.12}$$

An interesting mathematical concomittant of this simple behaviour is that the calculation of the discrete eigenvalue $\lambda_0(x^{\dagger})$ by first-order perturbation theory—taking the perturbation to be the truncated part of the simple relaxation kernel $K(x, y)$ for $x, y > x^{\dagger}$—proves to be exact. This result would appear to depend on the uniqueness of the discrete eigenvalue and seems unlikely to hold for more general models or for the complete Rayleigh problem with variable mass ratio.

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