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On the validity of the Brussels formalism in statistical mechanics

P V Coveney† and O Penrose‡

† Schlumberger Cambridge Research Laboratory, High Cross, Cambridge, CB3 0HG, UK

‡ Department of Mathematics, Heriot-Watt University, Riccarton, Edinburgh, EH14 4AS, UK

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Abstract. The so-called ‘Brussels’ approach to the derivation of kinetic equations usually proceeds by representing the Liouville operator L in the form $L_0 + \lambda L_1$ where λ is a perturbation parameter. It can be formulated in terms of an operator P (or a set of such operators) commuting with L_0 and projecting from a Hilbert space \mathcal{H} , spanned by all square-integrable phase-space densities or density matrices ρ , into a subspace \mathcal{H}_1 in which the reduced or kinetic description is to apply. For the case where \mathcal{H}_1 is finite-dimensional, we prove here two main results. (i) If the time-domain collision operator, defined by

$$\psi(t) = PL_1Q \exp(QLQ t)QL_1P$$

where $Q = 1 - P$, is bounded above in norm by a decreasing exponential function of $|t|$ and satisfies the condition that the Hermitian part of $\int_0^\infty e^{iyt}\psi(t) dt$ be invertible for all real y , then for sufficiently small positive λ the long-time asymptotic approach to equilibrium in the subspace \mathcal{H}_1 is an exponential decay or exponentially decaying oscillation and is correctly given by the Brussels perturbation method. (ii) If the norm of the collision operator decays to zero as $t \rightarrow \infty$ more slowly than any exponential then, regardless of the value of λ , the asymptotic behaviour does not have the exponential form predicted by the Brussels method.

1. Introduction

A central problem of non-equilibrium statistical mechanics is the derivation of kinetic equations: that is, an equation or set of equations giving the time evolution of some reduced description of the system. Most rigorous derivations of kinetic equations are valid only in the limiting case where some coupling parameter representing a strength of interaction becomes vanishingly small. A case in point is Lanford’s derivation of Boltzmann’s equation for a hard-sphere system [1], in which the small parameter is the diameter of one of the hard spheres.

In the late 1960s and early 1970s a group working in Brussels, notably Prigogine *et al* [2], pioneered an approach whose aim was to derive kinetic equations which were exact not just in a limiting case but over some finite range of values of the relevant coupling parameter. This approach has attracted the attention of a number of authors [3] and has been applied in fields outside statistical mechanics, including

the theory of dynamical systems [4] and the theory of decaying states in quantum mechanics [5]. The version of the method which we shall study here is based on the Nakajima–Zwanzig equation [6], an equation whose structure is similar to that of the desired kinetic equation, but differs from it in being non-autonomous: the equation contains a memory term so that the time rate of change of the reduced description depends on the reduced description at earlier times as well as at the time in question. The idea is to show that the long-time asymptotic behaviour of solutions of the Nakajima–Zwanzig equation is governed by an autonomous equation, without memory terms. This autonomous equation (equation (78)) in the present paper) is then the exact kinetic equation sought by the Brussels group.

A further strand in this thinking is the idea of subdynamics. The method gives a recipe for passing from any time-dependent ensemble density (by which we mean a phase-space density or, in the quantum case, a density matrix) ρ_t , for which the reduced description satisfies the kinetic equations only approximately, to a new ensemble density $\Pi\rho_t$ (really a measure rather than a density since it is not obviously non-negative) which, like ρ_t , satisfies Liouville's equation and for which the corresponding reduced description exactly satisfies the kinetic equation (78). The new measure $\Pi\rho_t$ is sometimes called the 'kinetic part' of ρ_t and the set of all possible $\Pi\rho_t$, a subspace of the linear space of all measures on phase space, is called the kinetic subspace. The kinetic subspace is invariant under the action of the Liouville operator, and the dynamical evolution that takes place in it is therefore described as 'subdynamics'. In many situations, such as the important cases of thermodynamic equilibrium and of non-equilibrium stationary states, it has been claimed [2] that the entire behaviour of the system lies within the kinetic subspace.

The Brussels theory relies heavily on two mathematical techniques. One is a perturbation method in which the Liouville operator L is regarded as being obtained by perturbing a simpler operator L_0 , for which the reduced description would be a complete representation of the time evolution, and everything is expanded in powers of a parameter measuring the size of this perturbation. The other is the use of analytic continuation in the complex frequency plane to approximate the long-time behaviour in terms of poles of the analytically continued resolvent of a related operator QLQ . These poles lead to exponentially decaying terms in the asymptotic time evolution. However, the manipulations used are usually formal rather than rigorous so that the reader may be left wondering, at best, whether the expansions used are sufficiently uniform in the time variable, to tell us anything useful about the long-time behaviour, and at worst whether they mean anything whatever.

The purpose of this paper is to look at a small part of the Brussels formalism from a mathematically rigorous point of view. We shall find that there are some conditions under which at least a part of the formalism rests on solid foundations; but on the other hand it is quite possible that these conditions cannot be satisfied for any realistic system. Indeed, a consequence of these conditions is that the long-time behaviour be an exponential decay; yet the widespread occurrence of 'long-time tails' in correlation functions [7], which decay with time more slowly than any exponential, indicates that there are many systems for which these conditions are not satisfied. It seems likely that such systems will not satisfy the conditions of validity for the Brussels method. For example, Dorfman and Cohen [8] have shown in the case of the density expansion for transport coefficients in imperfect gases that the terms giving rise to the long-time tails are the same ones as responsible for the divergence of the perturbation expansions.

2. Deriving the master equation

The fundamental object in the theory is the classical phase space density or quantum density matrix at time t , denoted ρ_t . We assume ρ_t to be square-integrable over phase space, so that it can be treated as an element in a Hilbert space \mathcal{H} , with inner product $(\rho, \sigma) = \int \bar{\rho}\sigma dx \cdots dq_N$ for classical mechanics, $\text{tr}(\rho^\dagger\sigma)$ for quantum mechanics. The Hilbert-space elements ρ_t for different times t are related by

$$\rho_t = U(t)\rho_0 \tag{1}$$

where the operators $U(t)$, acting in \mathcal{H} , are unitary and form a group with the multiplication law $U(t)U(s) = U(t+s)$. The generator L of this group, defined by

$$U(t) = \exp(Lt) \tag{2}$$

is an anti-Hermitian operator in the space \mathcal{H} . From (1) and (2) it follows that

$$\frac{d\rho_t}{dt} = L\rho_t \tag{3}$$

and so we can identify L as the Liouville operator.

Next one introduces a projection operator P , which projects from the full microscopic description associated with ρ_t to the reduced description where the kinetic equations are to apply. (In some applications of the Brussels method, e.g. [4] and [9], a family of such operators is used; for these applications, our P can be any member of this family, or a sum of such members.) An equation for the time evolution of $P\rho_t$ can be obtained by pre-multiplying both sides of (3) in turn by P and by the complementary projection operator Q defined by

$$Q = I - P \tag{4}$$

I being the unit operator. This gives a pair of differential equations

$$\frac{d}{dt}P\rho_t = PL\rho_t = PLP\rho_t + PLQ\rho_t \tag{5}$$

$$\frac{d}{dt}Q\rho_t = QL\rho_t = QLP\rho_t + QLQ\rho_t. \tag{6}$$

Solving for $Q\rho_t$ in (6) gives

$$Q\rho_t = \exp(tQL)Q\rho_0 + \int_0^t ds \exp[(t-s)QL]QLP\rho_s. \tag{7}$$

Insertion of this expression into (5) leads to the Nakajima–Zwanzig [6] equation which, since it holds for any ρ_0 , can be written (using (1)) as an operator equation for $U(t)$

$$\frac{d}{dt}PU(t) = PLPU(t) + PLQF(t)Q + \int_0^t ds PLQF(t-s)QLPU(s) \tag{8}$$

where we have defined

$$F(t) = \exp(tQLQ). \quad (9)$$

Equation (8) is subject to the initial condition

$$U(0) = 1. \quad (10)$$

In the perturbation method used by the Brussels group, the Liouville operator has the form

$$L = L_0 + \lambda L_1 \quad (11)$$

where L_0 and L_1 are both anti-Hermitian and λ is a small parameter, which we take here to be real so as to make L anti-Hermitian. The perturbing term λL_1 can be thought of as representing, for example, the effect of collisions. It is also assumed that

$$PL_0 = L_0P \quad (12)$$

so that if the system starts in the $P = 1$ subspace, then the unperturbed ($\lambda = 0$) time evolution does not take the system outside that subspace. It is usual to assume further that $PL_0 = L_0P = 0$ so that the time variation of the reduced description is due entirely to the small term λL_1 ; however, we shall not need this assumption here.

The kinetic equation is an evolution equation for the reduced phase-space density $P\rho_t$. To calculate the time evolution of $P\rho_t$ exactly with given initial conditions it is, by (1), necessary (and, if $Q\rho_0 = 0$, sufficient) to know the operator

$$V(t) = PU(t)P. \quad (13)$$

An integro-differential equation for $V(t)$ can be obtained by post-multiplying equation (8) by P and using the fact, which follows from (11), (4) and (12), that $PLQ = \lambda PL_1Q$ and $QLP = \lambda QL_1P$. The resulting equation, known in this context as the master equation, is

$$\frac{d}{dt}V(t) = PLPV(t) + \lambda^2 \int_0^t ds \psi(t-s)V(s) \quad (14)$$

with the initial condition

$$V(0) = P \quad (15)$$

and ψ (the so-called 'collision operator') defined by

$$\psi(t) = PL_1QF(t)QL_1P. \quad (16)$$

Note that ψ depends on λ as well as on t ; however our main results do not require any assumption as to the nature of this dependence.

To solve equation (14), define the Laplace transform of ψ as

$$\tilde{\psi}(p) = \int_0^\infty e^{-pt} \psi(t) dt \tag{17}$$

and $\tilde{V}(p)$ similarly. Taking the Laplace transform on both sides of (14) we obtain

$$p\tilde{V}(p) - P = PLP\tilde{V}(p) + \lambda^2\tilde{\psi}(p)\tilde{V}(p) \tag{18}$$

from which

$$\tilde{V}(p) = P[p - PLP - \lambda^2\tilde{\psi}(p)]^{-1}. \tag{19}$$

On taking the inverse Laplace transform we find

$$V(t) = \frac{1}{2\pi i} \int_{c-i\infty}^{c+i\infty} P[z - PLP - \lambda^2\tilde{\psi}(z)]^{-1} e^{zt} dz \quad (t > 0) \tag{20}$$

with c a real number chosen sufficiently large to ensure that the contour lies to the right of all singularities of the integrand in the complex z -plane.

To find the asymptotic behaviour of $V(t)$, one wishes to pick out those singularities within the integrand of equation (20) that provide the slowest decay when t is large. In this endeavour, the analytic properties of $\tilde{\psi}(z)$ are evidently of central concern.

3. Exponential time decay

In general, $V(t)$ is an operator. In this section and the next two, however, we restrict ourselves to the special case where the $P = 1$ eigenspace of P (which we shall call \mathcal{H}_1) is one-dimensional, so that $\psi(t)$ and $V(t)$ may be treated as scalar quantities. We shall also make the further simplification of assuming that

$$PLP = 0. \tag{21}$$

Equation (21) holds, for example, in the important case where ρ_t is a density matrix, $P\rho_t$ is the diagonal part of this matrix and $LP\rho_t$ is obtained by taking the commutator of $P\rho_t$ with the Hamiltonian operator. Later, in section 6, we shall generalize to the case when \mathcal{H}_1 is n -dimensional and where $PLP \neq 0$.

The following theorem, which is the central result of this paper, proves that if $|\psi(t)|$ is bounded by a decaying exponential then the dominant long-time behaviour of $V(t)$ agrees with the results of the Brussels group.

Theorem 1. Let $\psi(t)$ be a complex-valued function of t ($0 \leq t < \infty$) which is locally integrable and satisfies

$$|\psi(t)| < K \exp(-\alpha t) \quad \text{all } t > 0 \tag{22}$$

where K, α are positive constants. Let λ be a real number satisfying

$$\lambda^2 < \alpha^2 / 4K. \tag{23}$$

Then:

(i) The equation

$$z = \lambda^2 \tilde{\psi}(z) \quad (24)$$

with $\tilde{\psi}$ defined as in (17), has a unique solution in the half-plane $\text{Re } z > -\frac{1}{2}\alpha$ where Re indicates 'real part of'. We denote this solution by p_1 .

(ii) The one-dimensional version of equation (14) with $PLP = 0$ has a solution in the form

$$V(t) = W(t) + \hat{W}(t) \quad \text{all } t > 0 \quad (25)$$

with

$$W(t) = A \exp(p_1 t) \quad \text{all } t > 0 \quad (26)$$

and

$$|\hat{W}(t)| < K_1 \exp(-\frac{1}{2}\alpha t) \quad \text{all } t > 0 \quad (27)$$

where K_1 is a positive constant and A is defined by

$$A = \frac{1}{1 - \lambda^2 \tilde{\psi}'(p_1)} \quad (28)$$

the prime denoting a derivative. For large t the asymptotic form of $V(t)$ is therefore

$$V(t) \sim A \exp(p_1 t) \quad (t \rightarrow \infty) \quad (29)$$

with

$$-\frac{1}{2}\alpha < \text{Re } p_1. \quad (30)$$

(iii) The real part of p_1 is non-positive, so that $V(t)$ is bounded for large t . If

$$\text{Re } \tilde{\psi}(z) \neq 0 \quad \text{whenever } \text{Re } z = 0 \quad (31)$$

then the real part of p_1 is negative, so that $|V(t)|$ decays exponentially to zero as $t \rightarrow \infty$.

Proof. (i) Since $|\psi(t)|$ has an upper bound proportional to $\exp(-\alpha t)$, its Laplace transform $\tilde{\psi}(z)$ is, by (17), analytic in the open half-plane $\text{Re } z > -\alpha$. Thus, by (19) and (21), the only singularities of $\tilde{V}(z)$ in this open half-plane are the zeros of $z - \lambda^2 \tilde{\psi}(z)$. We apply Rouché's theorem to the function $z - \lambda^2 \tilde{\psi}(z)$ taken around a contour consisting of a large semicircle in the half-plane $\text{Re } z \geq -\frac{1}{2}\alpha$ with the mid-point of its straight side at the point $z = -\frac{1}{2}\alpha$. On this contour we have $|z| \geq \frac{1}{2}\alpha$ but, by (17), (22) and (23)

$$\lambda^2 |\tilde{\psi}(z)| \leq \lambda^2 \int_0^\infty e^{\alpha t/2} K e^{-\alpha t} dt = 2\lambda^2 K/\alpha < \frac{1}{2}\alpha. \quad (32)$$

So, by Rouché's theorem, the function $z - \lambda^2 \tilde{\psi}(z)$ has precisely one zero inside the half-plane, and this zero is the number p_1 defined in (24).

(ii) Deforming the contour in equation (20) and using (21) we get

$$V(t) = \frac{1}{2\pi i} \int_{-\alpha/2-i\infty}^{-\alpha/2+i\infty} \frac{e^{zt} dz}{z - \lambda^2 \tilde{\psi}(z)} + \frac{1}{2\pi i} \oint \frac{e^{zt} dz}{z - \lambda^2 \tilde{\psi}(z)} \quad (t > 0) \tag{33}$$

the second integral being taken around a small closed contour surrounding the pole at p_1 . The change in the path of integration at infinity is justified because of the lemma (theorem 13 in chapter II of [10]) that if the integral $f(z) = \int_0^\infty e^{-zt} d\mu(t)$ has an abscissa of convergence x_c then $f(x + iy) = o(|y|)$ as $|y| \rightarrow \infty$, uniformly in $x_c + \delta \leq x < \infty$ for any positive δ ; one need only take $\mu(t) = \int_0^t \psi(s) ds$ to see that the integrand in (20) has an upper bound of the form constant/(Im z) for large values of Im z , the imaginary part of z .

Defining the second integral in (33) to be $W(t)$ we have, by Cauchy's theorem of residues,

$$W(t) := \frac{1}{2\pi i} \oint \frac{e^{zt} dz}{z - \lambda^2 \tilde{\psi}(z)} = \frac{e^{p_1 t}}{1 - \lambda^2 \tilde{\psi}'(p_1)} \tag{34}$$

provided that $1 - \lambda^2 \tilde{\psi}'(p_1) \neq 0$. To check this last condition we note that, by (17), (22) and (23)

$$\lambda^2 |\tilde{\psi}'(p_1)| = \lambda^2 \left| \int_0^\infty e^{-p_1 t} \psi(t) t dt \right| \leq \lambda^2 4K/\alpha^2 < 1 \tag{35}$$

and so (26) and (28) are proved.

The other integral in (33) we define to be $\hat{W}(t)$, so that (25) holds automatically. To estimate $\hat{W}(t)$ we rewrite its definition as

$$\hat{W}(t) := \frac{1}{2\pi i} \int_{-\alpha/2-i\infty}^{-\alpha/2+i\infty} \left(\frac{1}{z} + \frac{\lambda^2 \tilde{\psi}(z)}{z(z - \lambda^2 \tilde{\psi}(z))} \right) e^{zt} dz. \tag{36}$$

Using the standard result

$$\frac{1}{2\pi i} \int_{-\alpha/2-i\infty}^{-\alpha/2+i\infty} \frac{e^{zt} dz}{z} = 0 \quad (t > 0) \tag{37}$$

and setting $z = -\frac{1}{2}\alpha + iy$ we obtain

$$\hat{W}(t) = e^{-\alpha t/2} \frac{1}{2\pi i} \int_{-\infty}^\infty \frac{\lambda^2 \tilde{\psi}(z) e^{iyt} dy}{(-\alpha/2 + iy)(-\alpha/2 + iy - \lambda^2 \tilde{\psi}(z))}. \tag{38}$$

It follows, by (17) and (23), that (27) holds, with

$$K_1 = \frac{1}{2\pi} \int_{-\infty}^\infty \frac{(2\lambda^2 K/\alpha) dy}{\sqrt{(\alpha^2/4 + y^2)} \{ \sqrt{(\alpha^2/4 + y^2)} - 2\lambda^2 K/\alpha \}}. \tag{39}$$

(iii) To prove that $\text{Re } p_1 \leq 0$ when (31) holds we use the result proved in appendix 1, which shows that equation (24) cannot be satisfied with $\text{Re } z > 0$. If in addition the condition (31) is satisfied then equation (24) cannot even be satisfied when $\text{Re } z = 0$, so that in that case we must have $\text{Re } p_1 < 0$. This completes the proof of theorem 1. □

The theorem shows that $V(t)$ is the sum of two parts. One of them, $\hat{W}(t)$, decays at least as fast as $e^{-\frac{1}{2}\alpha t}$ however small the value of λ is, but the other, $W(t)$, decays at a rate proportional to λ^2 . The condition (23) ensures that λ is small enough for this last term to dominate when t is large.

The implication of theorem 1 is that if the modulus of ψ is bounded above by a decreasing exponential, then the separation (equation (25)) used by the Brussels group holds in the $P \approx 1$ subspace \mathcal{H}_1 . There are discrete-time dynamical systems for which the function analogous to ψ can be shown to obey this sufficient condition, but we know of no continuous-time system for which this can be done.

In the work of the Brussels group a condition similar to (31), namely

$$\tilde{\psi}(0) \neq 0 \quad (40)$$

has been called the dissipativity condition [11]. If this condition is not satisfied, that is if $\tilde{\psi}(0) = 0$, then the relevant solution of equation (24) is $p_1 \approx 0$ and hence equation (26) shows that $V(t)$ approaches a constant for large t , so that at least part of the initial information contained in $P\rho_0$ is remembered by the system for all time. However, even with the condition (40) satisfied, $\tilde{\psi}(0)$ might be imaginary, in which case p_1 would be (to lowest order in λ) imaginary, $V(t)$ would oscillate sinusoidally for large t , and again some part of the initial information contained in $P\rho_0$ would be remembered by the system for all time. The name 'dissipativity condition' might better be given to equation (31) which, as our theorem shows, does guarantee dissipation, at least in the case considered here for which $PLP = 0$ and $\dim \mathcal{H}_1 = 1$.

The Brussels group introduced an operator $\Sigma(t)$ which is supposed to pick out the asymptotic behaviour of the system at large times [2]. In their notation, W and \hat{W} would be written $W(t) = P\Sigma(t)P$ and $\hat{W} = P\hat{\Sigma}(t)P$. However, we have no rigorous information about the behaviour of $\Sigma(t)$ beyond that given above concerning its restriction to the P subspace, namely $W(t)$.

4. Series expansions

In this section we show how some series expansions used by the Brussels group can be derived in a simple and rigorous way using complex variable methods. The theorem given here provides sufficient conditions for such series expansions to be valid. A principal requirement is that $\psi(t)$ have the exponential decay property used in theorem 1. If this requirement is not satisfied, the theorem in Section 5 makes it likely that the series expansions are not valid.

Theorem 2. Under the conditions of theorem 1 the propagator $W(t)$ defined in equation (26) has the following convergent series expansion:

$$W(t) = \sum_{n=0}^{\infty} \lambda^{2n} W_n(t) \quad (41)$$

where, for all t , we define

$$W_0(t) = 1 \quad (42)$$

$$W_n(t) = \frac{1}{n!} \int_0^{\infty} ds_1 \cdots \int_0^{\infty} ds_n (t - s_1 - \cdots - s_n)^n \psi(s_1) \cdots \psi(s_n).$$

The corresponding expansions for $V(t)$ and $\hat{W}(t)$ ($t \geq 0$) are

$$V(t) = 1 + \sum_{n=1}^{\infty} \frac{\lambda^{2n}}{n!} \int_0^{\infty} ds_1 \cdots \int_0^{\infty} ds_n (t - s_1 - \cdots - s_n)_+^n \psi(s_1) \cdots \psi(s_n) \quad (43)$$

$$\hat{W}(t) = - \sum_{n=1}^{\infty} \frac{\lambda^{2n}}{n!} \int_0^{\infty} ds_1 \cdots \int_0^{\infty} ds_n (t - s_1 - \cdots - s_n)_-^n \psi(s_1) \cdots \psi(s_n) \quad (44)$$

where, for any x , x_+ means x if $x \geq 0$ and 0 if $x < 0$, whilst x_- means x if $x \leq 0$ and 0 if $x > 0$ (so that $x^n = x_+^n + x_-^n$). It should be noted that, despite appearances, the expansions such as (41) are not power series expansions of the usual type, since the coefficients of λ^{2n} are themselves functions of λ .

Proof. We consider formula (41) first. Let λ satisfy the condition (23) as in theorem 1 and let the contour in equation (34) be a circle of radius $\frac{1}{2}\alpha$ centred at the origin. The definition (17) of $\tilde{\psi}$ and the condition (23) imply that $|\tilde{\psi}(z)| \leq 2K/\alpha$ on the contour; consequently we have

$$\begin{aligned} W(t) &= \frac{1}{2\pi i} \oint dz \frac{e^{zt}}{z - \lambda^2 \tilde{\psi}(z)} \\ &= \frac{1}{2\pi i} \oint dz e^{zt} \sum_{n=0}^{\infty} \lambda^{2n} \frac{\{\tilde{\psi}(z)\}^n}{z^{n+1}} \\ &= \sum_{n=0}^{\infty} \frac{\lambda^{2n}}{2\pi i} \oint dz e^{zt} \frac{\{\tilde{\psi}(z)\}^n}{z^{n+1}}. \end{aligned} \quad (45)$$

The third equality follows from the fact that the series inside the second integral is dominated by the series

$$\sum_n |\lambda^{2n}| (2K/\alpha)^n / (\frac{1}{2}\alpha)^{n+1} = \sum_n [\lambda^2 / (\alpha^2/4K)]^n / (\frac{1}{2}\alpha)$$

which converges due to (23). Thus, using Cauchy's formula for a derivative, we may write $W(t)$ in the form (41), convergent for $\lambda^2 < \alpha^2/4K$, where by (17)

$$W_0(t) = 1 \quad (\text{all } t) \quad (46)$$

$$\begin{aligned} W_n(t) &= \frac{1}{2\pi i} \oint dz e^{zt} \frac{\{\tilde{\psi}(z)\}^n}{z^{n+1}} \\ &= \frac{1}{n!} \left[\left(\frac{d}{dz} \right)^n e^{zt} \{\tilde{\psi}(z)\}^n \right]_{z=0} \\ &= \frac{1}{n!} \left[\left(\frac{d}{dz} \right)^n \int_0^{\infty} ds_1 \cdots \int_0^{\infty} ds_n e^{z(t-s_1-\cdots-s_n)} \psi(s_1) \cdots \psi(s_n) \right]_{z=0} \\ &= \frac{1}{n!} \int_0^{\infty} ds_1 \cdots \int_0^{\infty} ds_n (t - s_1 - \cdots - s_n)^n \psi(s_1) \cdots \psi(s_n). \end{aligned}$$

Now we consider the series for $V(t)$, equation (43). The multiple integral in the n th term in the series for $V(t)$, call it $V_n(t)$, is bounded above, when $t > 0$, by

$$\begin{aligned}
 & \int_0^\infty ds_1 \cdots \int_0^\infty ds_n (t - s_1 - \cdots - s_n)_+^n |\psi(s_1)| \cdots |\psi(s_n)| \\
 & \leq \int_0^\infty ds_1 \cdots \int_0^\infty ds_n t^n |\psi(s_1)| \cdots |\psi(s_n)| \\
 & = t^n \left(\int_0^\infty ds |\psi(s)| \right)^n.
 \end{aligned}
 \tag{47}$$

Hence the series (43) for $V(t)$ is majorized by the series for $\exp(|\lambda|^2 t \int_0^\infty ds |\psi(s)|)$, and therefore converges by virtue of the bound (22) on $\psi(t)$, uniformly on any compact interval of the positive t -axis. Then it may be checked, using term-by-term differentiation, that the series for $V(t)$ satisfies the master equation (14) and its boundary condition $V(0) = 1$.

Finally, to prove the series (44) for $\hat{W}(t)$ we substitute (41) and (43) into (25). This completes the proof of theorem 2. □

The expansions in equations (41)–(44) are exactly the same as ones used by the Brussels group [2, 9].

5. Non-exponential time evolution

In this section, we consider the case where the asymptotic time evolution decays more slowly than any exponential function. The following theorem gives a sufficient condition for such a decay situation.

Theorem 3. Let ψ be a locally integrable complex-valued function on $[0, \infty)$ which decays to zero more slowly than any exponential, so that

$$\lim_{t \rightarrow \infty} \psi(t) = 0
 \tag{48}$$

whilst for any positive α , however small, we have

$$\psi(t)e^{\alpha t} \quad \text{unbounded as } t \rightarrow \infty.
 \tag{49}$$

Then the solution $V(t)$ of the one-dimensional version of equation (14), provided that its fourth derivative exists and is of bounded variation, is not bounded on the positive t -axis by any decreasing exponential. This result holds even if PLP is non-zero.

Proof. Suppose, to the contrary, that $V(t)$ were bounded in absolute value by some decreasing exponential of the form $C \exp(-\gamma t)$ with $\gamma > 0$ and C a positive constant. Then its Laplace transform $\tilde{V}(p)$ would be analytic in the open half-plane $\text{Re } p > -\gamma$ and hence, by (19), the function $\tilde{\psi}(p)$ would be meromorphic for $\text{Re } p > -\gamma$. Consequently, by deforming the contour in the inverse transform formula giving $\psi(t)$ in terms of $\tilde{\psi}(p)$, we would have

$$\begin{aligned}
 \psi(t) &= \frac{1}{2\pi i} \int_{-\gamma_1 - i\infty}^{-\gamma_1 + i\infty} e^{zt} \lambda^{-2} \left(z - PLP - \frac{1}{\tilde{V}(z)} \right) dz + \frac{1}{2\pi i} \\
 &\times \{ \text{sum of residues at poles of integrand in } \text{Re } z > -\gamma_1 \}
 \end{aligned}
 \tag{50}$$

where γ_1 is a positive number less than γ . This last step is justified by the lemma referred to in the proof of theorem 1, this time applied with $\mu(t) = V''''(t)$ so that after five partial integrations and use of the facts (deducible from equations (13) and (2) restricted to \mathcal{H}_1) that $V(0) = 1$ and $V'(0) = PLP$ the lemma gives

$$(x + iy)\tilde{V}(x + iy) = 1 + \frac{PLP}{(x + iy)} + \frac{V''(0)}{(x + iy)^2} + \frac{V'''(0)}{(x + iy)^3} + o\left(\frac{1}{|y|^3}\right) \quad (51)$$

uniformly in $-\gamma_1 < x < \infty$. From this version of the lemma it follows that the integrand in (50) is uniformly small for large $|y|$ so that the change in the path of integration at infinity is justified, and also that $1/V(x + iy) \sim (x + iy) - PLP + o(1)$ for large $|y|$ so that the poles mentioned in (50) are confined to some finite part of the strip $-\gamma_1 < \text{Re } z \leq 0$ and are therefore finite in number.

By an argument similar to the one starting with equation (36) in the proof of theorem 1, using the estimate (51), the first term on the right of (50) decays as $t \rightarrow \infty$ faster than any exponential $e^{-\gamma_2 t}$ with $0 < \gamma_2 < \gamma_1$, and the second is a sum of terms of the form $e^{p_i t} \times (\text{polynomial in } t)$, where p_i is the position of the relevant pole. But such a form for $\psi(t)$ is incompatible with at least one of the conditions (48), (49); so our original supposition must be false, and the theorem is proved. \square

As a corollary, theorem 3 implies that if $|\psi(t)|$ falls off more slowly than exponentially and $V(t)$ is smooth enough then the sum of the convergent power series (43) for $V(t)$ does not decay exponentially with increasing t . In this case there is no guarantee that the integrals defining the series expansions (42) and (44) for $W(t)$ and $\tilde{W}(t)$ exist, and even if they do there is no guarantee that the expansions will converge.

6. The case where \mathcal{H}_1 is n -dimensional

In the foregoing work we made the assumption that the subspace \mathcal{H}_1 picked out by the projection operator P was one-dimensional. Here we extend the analysis to deal with the case where this subspace is n -dimensional (n being finite) and at the same time drop the restriction $PLP = 0$ (equation (21)).

As before, the starting point is equation (14), but now $F(t)$ and $\psi(t)$ in equations (16) and (9) are operators in \mathcal{H}_1 . They can be represented (relative to some arbitrary basis in \mathcal{H}_1) as $n \times n$ matrices. Then $V(t) = PU(t)P$ in equation (13) is also represented by an $n \times n$ matrix, and equation (14) can be interpreted as a matrix equation subject to the boundary condition $V(0) = I_n$, where I_n is the unit $n \times n$ matrix. Taking the Laplace transform of the matrix version of equation (14) we have, using (11)

$$\tilde{V}(p) = \{pI_n - PLP - \lambda^2 \tilde{\psi}(p)\}^{-1} \quad (52)$$

and so the solution to the master equation (14) is now

$$V(t) = \frac{1}{2\pi i} \int_{c-i\infty}^{c+i\infty} dz e^{zt} \{zI_n - PLP - \lambda^2 \tilde{\psi}(z)\}^{-1} \quad (53)$$

with c sufficiently large.

We use the standard Hilbert-space operator norm in \mathcal{H}_1 , which is given by

$$\|\psi\|^2 = (\text{largest eigenvalue of } \psi^\dagger\psi) \tag{54}$$

so that

$$n^{-1}\text{tr}(\psi^\dagger\psi) \leq \|\psi\|^2 \leq \text{tr}(\psi^\dagger\psi). \tag{55}$$

Theorem 4. Let $\psi(t)$ be a function of $t \in [0, \infty)$ whose values are operators in \mathcal{H}_1 satisfying

$$\|\psi(t)\| < K \exp(-\alpha t) \quad \text{all } t > 0 \tag{56}$$

where K and α are positive constants, and let λ be a real number satisfying

$$\lambda^2 < \frac{1}{4K} \alpha^2 \sin^{-1} \frac{\pi}{2n}. \tag{57}$$

Define p_1, p_2, \dots to be the solutions of

$$\det\{zI_n - PLP - \lambda^2\tilde{\psi}(z)\} = 0 \tag{58}$$

that lie in the half-plane $\text{Re } z \geq -\frac{1}{2}\alpha$, and suppose that the n eigenvalues of the matrix $PLP + \lambda^2\tilde{\psi}(z)$ are distinct for all z in some neighbourhood of the point set $\{p_1, p_2, \dots\}$. Then

- (i) the number of points in the set $\{p_1, p_2, \dots\}$ (allowing for multiplicities) is n .
- (ii) The matrix version of equation (14) has a solution in the form

$$V(t) = W(t) + \hat{W}(t) \quad \text{all } t > 0 \tag{59}$$

with

$$\|\hat{W}(t)\| < K_1 \exp(-\frac{1}{2}\alpha t) \quad \text{all } t > 0 \tag{60}$$

where K_1 is a positive constant and, if the points p_1, \dots, p_n are distinct,

$$W(t) = \sum_{i=1}^n c_i e^{p_i t} |u_i\rangle \langle v_i| \tag{61}$$

where $|u_i\rangle$ is a non-zero $n \times 1$ column vector satisfying the equation

$$\{p_i I_n - PLP - \lambda^2\tilde{\psi}(p_i)\} |u_i\rangle = 0 \tag{62}$$

$\langle v_i|$ is a non-zero $1 \times n$ row vector satisfying

$$\langle v_i| \{p_i I_n - PLP - \lambda^2\tilde{\psi}(p_i)\} = 0 \tag{63}$$

and c_i is the number defined by

$$c_i = \frac{1}{\langle v_i| \{I_n - \lambda^2\tilde{\psi}'(p_i)\} |u_i\rangle}. \tag{64}$$

(iii) The real parts of the numbers $\{p_1, \dots, p_n\}$ are non-positive. If the Hermitian part of the operator $\tilde{\psi}(z)$ is invertible for all purely imaginary values of z then the real parts of $\{p_1, \dots, p_n\}$ are negative.

Proof. (i) We apply Rouché's theorem to the function

$$\begin{aligned}
 f(z) &= \det\{zI_n - PLP - \lambda^2\tilde{\psi}(z)\} \\
 &= \det\{zI_n - PLP\} \det\{I_n - (zI_n - PLP)^{-1}\lambda^2\tilde{\psi}(z)\} \quad (65)
 \end{aligned}$$

taken round the semicircular contour defined in the proof of theorem 1. Since PLP has only a finite number of eigenvalues, all of them purely imaginary, we can make the semicircle big enough to ensure that the distance from a point on the contour to an eigenvalue of PLP is always at least $\frac{1}{2}\alpha$, and hence that

$$\|(zI_n - PLP)^{-1}\| \leq (\frac{1}{2}\alpha)^{-1} \quad (66)$$

for all z on the contour. We also have, as in the proof of theorem 1,

$$\|\tilde{\psi}(z)\| \leq 2K/\alpha \quad (67)$$

and hence, by (66), (57) and (67)

$$\|(zI_n - PLP)^{-1}\lambda^2\tilde{\psi}(z)\| < \sin^{-1}(\pi/2n). \quad (68)$$

It follows that each one of the eigenvalues of $\{I_n - (zI_n - PLP)^{-1}\lambda^2\tilde{\psi}(z)\}$ lies within a circle in the complex plane having centre 1 and radius $\sin^{-1}(\pi/2n)$, so that the eigenvalue cannot vanish and its argument is between $-\pi/2n$ and $\pi/2n$; consequently $\det\{I_n - (zI_n - PLP)^{-1}\lambda^2\tilde{\psi}(z)\}$ also cannot vanish and its argument is between $-\pi/2$ and $\pi/2$. As z goes all the way around the contour the total change in $\arg \det\{I_n - (zI_n - PLP)^{-1}\lambda^2\tilde{\psi}(z)\}$ must therefore be zero and so, by (65), $f(z)$ and $\det\{zI_n - PLP\}$ have the same number of zeros inside the contour. The zeros of $\det\{zI_n - PLP\}$, being the eigenvalues of PLP , all lie inside the contour and are n in number; so $f(z)$ has n zeros inside the contour and by definition these are the numbers p_1, \dots, p_n .

(ii) Deforming the contour in equation (53), we have

$$V(t) = \frac{1}{2\pi i} \int_{-\alpha/2-i\infty}^{-\alpha/2+i\infty} e^{zt} dz \{zI_n - PLP - \lambda^2\tilde{\psi}(z)\}^{-1} + W(t) \quad (69)$$

where the matrix function $W(t)$, which provides the asymptotic form of $V(t)$, is given (since we are assuming p_1, \dots, p_n to be distinct) by

$$W(t) = \sum_{i=1}^n \left[\text{residue at } p_i \text{ of } e^{zt} \{zI_n - PLP - \lambda^2\tilde{\psi}(z)\}^{-1} \right]. \quad (70)$$

The proof of formula (61), which follows from (70), is given in appendix 2. The proof of formula (60) is closely analogous to that of the corresponding formula (27) in theorem 1; there is no need to repeat the details.

(iii) By the definition (58) of p_i , there is a non-zero vector φ in \mathcal{H}_1 with the property

$$(\varphi, \{p_i I_n - PLP - \lambda^2\tilde{\psi}(p_i)\}\varphi) = 0. \quad (71)$$

Taking the real part we find, since PLP is anti-Hermitian, that

$$(\varphi, \varphi) \operatorname{Re} p_i - \lambda^2 \operatorname{Re} (\varphi, \tilde{\psi}(p_i)\varphi) = 0 \tag{72}$$

By the result in appendix 1, the expression $\{(\varphi, \varphi) \operatorname{Re} p - \lambda^2 \operatorname{Re} (\varphi, \tilde{\psi}(p)\varphi)\}$ is certainly positive if $\operatorname{Re} p$ is positive, and so $\operatorname{Re} p_i$ cannot be positive. If in addition the Hermitian part of $\tilde{\psi}(p)$ is non-singular for all purely imaginary p then, since a non-singular non-positive definite matrix must be negative definite, the Hermitian part of $\tilde{\psi}(p)$ is negative definite for purely imaginary p . Consequently in this case the expression

$$(\varphi, \varphi) \operatorname{Re} p - \lambda^2 \operatorname{Re} (\varphi, \tilde{\psi}(p)\varphi)$$

must be positive even when $\operatorname{Re} p = 0$ and so, by (72), $\operatorname{Re} p_i$ cannot even be zero. This completes the proof of theorem 4. \square

Even if the eigenvalues of $PLP + \lambda^2 \tilde{\psi}(z)$ are not distinct, equation (61) still holds provided that the matrices $p_i I_n - PLP - \lambda^2 \tilde{\psi}(p_i)$ ($i = 1, \dots, n$) all have rank $n - 1$. Some other cases are discussed briefly in appendix 2.

As in theorem 1 these results show that the asymptotic behaviour of $V(t)$ at large times is dominated by $W(t)$ and is therefore a sum of decaying exponentials and damped oscillations as given by (61).

Equation (61), in a slightly different form, was first given by Grecos *et al* [12].

Theorem 3 likewise generalizes to the case where \mathcal{H}_1 is n -dimensional: if $\|\psi(t)\|$ decays to zero more slowly than any decreasing exponential, then $\|V(t)\|$ is not bounded on the positive real axis by any decreasing exponential. The proof is a direct analogue of the one given in section 5 for the $n = 1$ case.

7. Asymptotic evolution equations

In the Brussels formalism, $W(t)$ is an important operator; as noted earlier, authors from this group write it as $P\Sigma(t)P$ since it is the projection into \mathcal{H}_1 of an operator $\Sigma(t)$ which when applied to the initial phase-space density ρ_0 is supposed to give the so-called 'kinetic' (long-time asymptotic) part of the complete phase space density ρ_t . The operator Σ is then used to construct the kinetic equation governing the approach to equilibrium [2].

To compare our results with those of the Brussels group, we can derive an evolution equation for $W(t)$ by differentiating equation (61) with respect to t , using equations (62), (17) and finally (61) again. This procedure gives, assuming for simplicity that $PLP = 0$ (equation (21))

$$\begin{aligned} \frac{d}{dt} W(t) &= \sum_{i=1}^n p_i e^{p_i t} |u_i\rangle c_i \langle v_i| \\ &= \sum_{i=1}^n \lambda^2 \tilde{\psi}(p_i) e^{p_i t} c_i |u_i\rangle \langle v_i| \\ &= \sum_{i=1}^n \lambda^2 \int_0^\infty e^{p_i(t-s)} \psi(s) ds c_i |u_i\rangle \langle v_i| \\ &= \lambda^2 \int_0^\infty \psi(s) W(t-s) ds. \end{aligned} \tag{73}$$

This result should be compared with the master equation (14) for the full evolution operator $V(t)$. Equation (73) coincides exactly with equation (74) of [9], which was obtained by the methods of the Brussels group.

Another operator from the Brussels formalism whose existence we may establish from the foregoing discussion is the so-called 'kinetic operator' [2, 9]. This operator, which the Brussels group denote by $P\Gamma P$, is defined by the requirement that it satisfy (in the case $PLP = 0$)

$$P\Gamma P = \lambda^2 \int_0^\infty ds \psi(s) e^{-sP\Gamma P}. \tag{74}$$

From this definition it follows by differentiation that

$$\frac{d}{dt} [e^{tP\Gamma P} W(0)] = P\Gamma P e^{tP\Gamma P} W(0) = \lambda^2 \int_0^\infty ds \psi(s) e^{(t-s)P\Gamma P} W(0). \tag{75}$$

Comparison with equation (73) shows that both $W(t)$ and $e^{tP\Gamma P} W(0)$ satisfy the same evolution equation and are the same at $t = 0$. Hence

$$W(t) = e^{tP\Gamma P} W(0). \tag{76}$$

From this it follows that

$$\frac{dW(t)}{dt} = P\Gamma PW(t). \tag{77}$$

If ρ_0 is chosen to lie in \mathcal{H}_1 then $W(t)\rho_0$, which according to (13) and (29) is the asymptotic form of $P\rho(t)$, will satisfy the equation

$$\frac{d}{dt} W(t)\rho_0 = P\Gamma PW(t)\rho_0 \tag{78}$$

which is the autonomous kinetic equation sought by the Brussels school, restricted to the subspace \mathcal{H}_1 .

It can be verified, using first equation (62) (with $PLP = 0$) and then (17), that a matrix satisfying equation (74) is

$$P\Gamma P = \sum_{i=1}^n p_i |u_i\rangle \langle u_i^*| \tag{79}$$

where $\langle u_i^*|$ denotes the element associated to $|u_i\rangle$ (as defined in (62)) in the corresponding dual basis, the set of row vectors satisfying

$$\langle u_i^*|u_j\rangle = \delta_{ij}. \tag{80}$$

Such a dual basis always exists, and is unique, provided the column vectors $|u_i\rangle$ are linearly independent. A sufficient condition for these vectors to be linearly independent at small λ (remembering that $PLP = 0$ here), is that the matrix $\tilde{\psi}(0)$ depend continuously on λ and have distinct eigenvalues when $\lambda = 0$; for then equation (62) with $PLP = 0$ shows that in the limit $\lambda \rightarrow 0$ the vectors $|u_i\rangle$ approach the eigenvectors of $\tilde{\psi}(0)$, and the latter are linearly independent if the eigenvalues of $\tilde{\psi}(0)$ are distinct [13]. The linear independence of the vectors $|u_i\rangle$ when λ is sufficiently small then follows by continuity.

Equation (79) was first given in [12].

8. Discussion

In this paper, we have made a mathematically rigorous study of some aspects of the long-time evolution governed by Liouville's equation. Our results establish that, if the norm of the collision operator $\psi(t)$ in the master equation is bounded above by a decreasing exponential, and if the dimension of the $P = 1$ subspace is finite, then the Brussels formalism does indeed pick out the most slowly varying part $W(t)$ of the complete solution; moreover, the series expansion for this component converges for sufficiently small values of the perturbation parameter λ . However, if these conditions on $\psi(t)$ are not met, the Brussels method is unlikely to work.

Some members of the Brussels group have attached much significance to exponential time decay in their writings, particularly in the context of the decays of unstable particles in quantum mechanics [14], although more recently it has been stated [4] that long-time tails (non-exponential decay) can be obtained from the formalism. Our work shows that exponential decay both of the collision operator and also in the approach to equilibrium are indeed characteristic features of situations where the Brussels formalism works but not necessarily of the behaviour of real physical systems. If it is indeed, as stated by Petrosky and Hasegawa [4], that long-time tails can be obtained from the Brussels formalism, a minimum requirement appears to be that \mathcal{H}_1 be infinite-dimensional.

Although we have followed the standard convention of the Brussels group in taking ρ_t to be the phase space density (or density matrix), there is a lot to be said for taking it instead to be the difference between the total phase-space density and the equilibrium phase-space density. In that case \mathcal{H} would denote not the entire Hilbert space spanned by the square-integrable phase-space densities, but the subspace of that Hilbert space consisting of square-integrable functions that are orthogonal to the equilibrium phase-space density or densities. The resulting theorems hold just as before, and the prospects for satisfying the conditions for the theory to apply are considerably brighter.

It would be desirable to try to find model systems which would satisfy the requirement on $\|\psi(t)\|$ of exponential decrease. From the definition of $\psi(t)$, it is clear that this property is related to the ergodic properties of QLQ . One may anticipate that mixing will be a minimum requirement; in fact, it has previously been conjectured (but not proved) that the dynamical system should be an Anosov flow [15].

For systems of particles with attractive interactions, it seems unlikely that the Anosov property will hold. Indeed, by the KAM theorem, we know that for weak attractive interactions the invariant subspaces of the Liouville operator depend in a very complicated and non-analytic way on the strength of the interaction. In such cases, it seems very unlikely that one can describe the long-time behaviour by means of an expansion in powers of a perturbation parameter and if so then the Brussels method would not be applicable.

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Appendix 1. Proof that the Hermitian part of $\tilde{\psi}(z)$ is non-positive definite when $\text{Re } z$ is non-negative.

We prove that, regardless of the dimension of \mathcal{H}_1 providing only that it is infinite, the Hermitian part of $\tilde{\psi}(z)$ is non-positive definite for any complex z with positive real part, and that it is also non-positive definite when z is purely imaginary if $\|\psi(t)\|$ satisfies the upper bound condition (56). This is equivalent to showing that $\text{Re}(\varphi, \tilde{\psi}(z)\varphi) \leq 0$ for all $\varphi \in \mathcal{H}_1$ and all z with $\text{Re } z \geq 0$. Using the definition (17) of the Laplace transform, the definition of $\psi(t)$ (equations (16) and (9)), and the fact that L is anti-Hermitian, this matrix element can be written

$$\begin{aligned} (\varphi, \tilde{\psi}(z)\varphi) &= (\varphi, PL_1Q \int_0^\infty dt e^{-(z-QLQ)t} QL_1P\varphi) \\ &= -(QL_1P\varphi, (z-QLQ)^{-1}QL_1P\varphi) \\ &= -\int_{-\infty}^\infty \frac{1}{x+iy-i\eta} (QL_1P\varphi, dE(\eta)QL_1P\varphi) \\ &= -\int_{-\infty}^\infty (QL_1P\varphi, dE(\eta)QL_1P\varphi) \left(\frac{x-i(y-\eta)}{x^2+(y-\eta)^2} \right) \end{aligned} \tag{81}$$

where $E(\eta)$ is the spectral resolution of the identity for the (Hermitian) operator $-iQLQ$ (so that the eigenvalues of QLQ are $i\eta$ with η real), and x, y are, respectively, the real and imaginary parts of z . Thus we have

$$-\text{Re}(\varphi, \tilde{\psi}(z)\varphi) = \int_{-\infty}^\infty (QL_1P\varphi, dE(\eta)QL_1P\varphi) \left(\frac{x}{x^2+(y-\eta)^2} \right). \tag{82}$$

For $x > 0$ the right-hand side is non-negative because $dE(\eta)$ is a projection; therefore $\text{Re}(\varphi, \tilde{\psi}(z)\varphi)$ is non-positive. If $\|\psi(t)\|$ satisfies the upper bound condition (56), then $(\varphi, \tilde{\psi}(z)\varphi)$, being analytic in z for $x > -\frac{1}{2}\alpha$ is continuous at $x = 0$ and therefore $\text{Re}(\varphi, \tilde{\psi}(z)\varphi)$ is non-positive for $x = 0$. QED.

In the limit $z \rightarrow iy$, the right-hand side of (82) becomes, formally

$$\int_{-\infty}^\infty (QL_1P\varphi, dE(\eta)QL_1P\varphi) \pi\delta(\eta) = \pi \frac{d}{d\eta} [(QL_1P\varphi, E(\eta)QL_1P\varphi)]_{\eta=y}. \tag{83}$$

Hence $\text{Re}(\varphi, \tilde{\psi}(iy)\varphi)$ is negative unless $(d/d\eta)[(QL_1P\varphi, E(\eta)QL_1P\varphi)]_{\eta=y} = 0$. In particular, if the spectrum of QLQ is continuous near $\eta = y$, then there will in general be some φ for which $\text{Re}(\varphi, \tilde{\psi}(iy)\varphi) < 0$.

Appendix 2. Proof of equation (61).

Define

$$G(z) = PLP + \lambda^2\tilde{\psi}(z). \tag{84}$$

The definition (58) of p_i is equivalent to the statement that p_i is an eigenvalue of $G(p_i)$. In accordance with our hypothesis that the eigenvalues of $G(z)$ are distinct,

there is a non-singular matrix $M(z)$, whose columns are the right eigenvectors of $G(z)$, such that the matrix $D(z)$ defined by

$$D(z) = M(z)^{-1}G(z)M(z) \tag{85}$$

is diagonal and has all its diagonal elements different. Hence there is for each p_i a nonsingular matrix $M(p_i)$ such that

$$D(p_i) = M(p_i)^{-1}G(p_i)M(p_i) \tag{86}$$

is also diagonal, and hence the diagonal matrix

$$I_n p_i - D(p_i) = M(p_i)^{-1}[I_n p_i - G(p_i)]M(p_i) \tag{87}$$

has precisely one zero element on the diagonal. Let this be the k th diagonal element; denote by $|e_k\rangle$ the column matrix with a 1 in the k th diagonal position and zeros everywhere else, and by $\langle e_k|$ the analogous row matrix.

To calculate the residues in equation (70), we need the inverse of the matrix

$$I_n z - D(z) = I_n p_i - D(p_i) + (z - p_i)(I_n - D'(p_i)) + O((z - p_i)^2). \tag{88}$$

On the diagonal, all elements of this matrix are $O(1)$ as $z \rightarrow p_i$ except the k th, which is $(z - p_i)\langle e_k|[I_n - D'(p_i)]|e_k\rangle + O((z - p_i)^2)$. Off the diagonal all elements are at most $O(z - p_i)$. Calculating the inverse matrix by means of cofactors, we find that

$$[I_n z - D(z)]^{-1} = |e_k\rangle \frac{(z - p_i)^{-1}}{\langle e_k|[I_n - D'(p_i)]|e_k\rangle} \langle e_k| + O(1) \tag{89}$$

provided that the denominator, which is the (k, k) element of $(I_n - D'(p_i))$, is non-zero. This denominator can be written, using (86), as

$$\begin{aligned} 1 - \frac{d}{dz} \langle e_k|D(z)|e_k\rangle \Big|_{z=p_i} &= 1 - \frac{d}{dz} \langle e_k|M(z)^{-1}G(z)M(z)|e_k\rangle \Big|_{z=p_i} && \text{by (86)} \\ &= 1 - \lambda^2 \langle e_k|M(z)^{-1}\tilde{\psi}'(z)M(z)|e_k\rangle \Big|_{z=p_i} && \text{by (84)} \end{aligned} \tag{90}$$

the terms arising from differentiation of $M(z)^{-1}$ and $M(z)$ giving no contribution because

$$\begin{aligned} &\left\langle e_k \left| \frac{d}{dz} [M(z)^{-1}]G(z)M(z) \right| e_k \right\rangle \Big|_{z=p_i} + \left\langle e_k \left| M(z)^{-1}G(z) \frac{d}{dz} M(z) \right| e_k \right\rangle \Big|_{z=p_i} \\ &= \left\langle e_k \left| \left[\frac{d}{dz} M(z)^{-1} \right] M(z) \right| e_k \right\rangle \Big|_{z=p_i} + p_i \left\langle e_k \left| M(z)^{-1} \frac{d}{dz} M(z) \right| e_k \right\rangle \Big|_{z=p_i} \\ &= p_i \frac{d}{dz} \langle e_k|M(z)^{-1}M(z)|e_k\rangle \Big|_{z=p_i} = 0. \end{aligned} \tag{91}$$

To find the residue required for the evaluation of (70) we multiply (89) on the left by $M(p_i)$ and on the right by $M(p_i)^{-1}$, and then use (90) and (87), obtaining

$$[I_n z - G(z)]^{-1} = M(p_i)|e_k \frac{(z - p_i)^{-1}}{\langle e_k | M(p_i)^{-1} [I_n - \lambda^2 \tilde{\psi}'(p_i)] M(p_i) | e_k \rangle} \langle e_k | M(p_i)^{-1} + O(1). \tag{92}$$

Since $M(p_i)|e_k$, the k th column of the matrix $M(p_i)$, is proportional to the column matrix $|u_i$ defined in (62) and $\langle e_k | M(p_i)^{-1}$ is proportional to $\langle v_i |$ defined in (63), the result (61) follows. □

The above calculation works in some cases where $G(z)$ does not have distinct eigenvalues, provided the numbers p_i are all distinct and each p_i is a non-degenerate eigenvalue of the corresponding $G(p_i)$, so that all the diagonal matrices $I_n p_i - D(p_i)$ have but a single zero on the diagonal—i.e. they are of rank $n - 1$. The method can be generalized to the case where some or all of these matrices have rank less than $n - 1$, provided they can still be diagonalized; in this case the formula for the residue involves the inverse of the diagonal submatrix of $I_n - D'(p_i)$ whose diagonal elements are in the same places as the zero diagonal elements of $I_n p_i - D(p_i)$, but the formula is not worth giving here.

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