

NIELS JOERGEN KOKHOLM

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# Spectral Analysis of Perturbed Multiplication Operators Occurring in Polymerization Chemistry.

NIELS JØRGEN KOKHOLM

Copenhagen University Mathematics Department

**0. Introduction.** We consider an evolution equation in  $L^1(\mathbf{R}_+)$  with a generator which is the sum of a multiplication operator and an integral operator. Existence and uniqueness for all times and initial values are proved together with an asymptotic formula for the solution as  $t \rightarrow \infty$ . The proof of the large time asymptotics is partly based on the study of a selfadjoint operator in  $L^2(\mathbf{R}_+)$ , which is the sum of a multiplication operator and an integral operator, whose kernel is the Green's function of a second order Sturm Liouville problem. The operators in  $L^1$  and  $L^2$  are related by conjugation with a multiplication operator. The operator in  $L^2$  has purely absolutely continuous spectrum  $[0, \infty[$  and infinitely many negative eigenvalues converging to 0 with very unusual asymptotics.

**1. An evolution equation in  $L^1(\mathbf{R}_+)$ .** In a paper on a kinetic model for polymerization in which the polymer molecules are built up of identical units, Thor A. Bak and Lu Binglin, [B-L], proposed the study of the following initial value problem

$$\begin{cases} \frac{\partial P}{\partial t}(x, t) = -(x+2)P(x, t) + 2 \int_x^\infty x/y P(y, t) dy + 2 \int_0^x e^{y-x} P(y, t) dy, & x, t > 0, \\ P(\cdot, 0) = P_0 \in \mathcal{P} = \{f \geq 0 \mid \int_0^\infty f(y) dy = 1\} & \text{is a given probability distribution.} \end{cases}$$

Here  $P(x, t)$  is the probability distribution, at time  $t$ , for the length of the polymer molecule which contains a particular (say marked) unit. We describe some of the results obtained in [Ko1] (cf. also [Ko2]).

First of all, we have existence and uniqueness for all times if we demand that

- (1)  $P(\cdot, t) \in L^1(\mathbf{R}_+)$  is continuous for  $t \geq 0$ , differentiable for  $t > 0$ ,
- (2)  $\int_0^\infty x|P(x, t)| dx < \infty$  for  $t > 0$ .

This time development preserves the set of probability distributions  $\mathcal{P}$  (as it should for chemical reasons). The proof of these facts goes as follows, the solution will be given by a contraction semigroup  $P(\cdot, t) = e^{-tA}P_0$ . Write the equation as  $\dot{P} = -A_1P$ , where  $A_1 = x + 2 - A_f - A_c$  and

$$A_f P(x) = 2 \int_x^\infty x/y P(y) dy, \quad A_c P(x) = 2 \int_0^x e^{y-x} P(y) dy.$$

When  $P_0 \in L^1_{comp}(\mathbf{R}_+)$ , the integrable functions with compact support, the exponential series for  $T(t)P_0 := e^{-t(x-A_f)}P_0$  converges and by the Trotter product formula,

$$T(t)P_0 = \sum_{n=0}^{\infty} \frac{1}{n!} (t(A_f - x))^n P_0 = \lim_{n \rightarrow \infty} (e^{-tx/n} e^{tA_f/n})^n P_0.$$

It follows easily that  $T(t)$  preserves  $\mathcal{P} \cap L^1_{comp}(\mathbf{R}_+)$ , so  $T(t)$  extends to a contraction semigroup preserving  $\mathcal{P}$ . The generator of  $T(t)$  is a restriction of  $x - A_f$ , namely the closure of the restriction to  $L^1_{comp}(\mathbf{R}_+)$ . Since  $2 - A_c$  is bounded in  $L^1(\mathbf{R}_+)$  and  $e^{-t(2-A_c)}$  preserves  $\mathcal{P}$ , the theory of perturbation of contraction semigroups gives a contraction semigroup  $e^{-tA}P_0 = \lim_{n \rightarrow \infty} (T(t/n)e^{-t(2-A_c)/n})^n P_0$  preserving  $\mathcal{P}$  and with the generator  $A$  equal to a restriction of  $A_1$ , namely the closure of  $A_1$  restricted to  $L^1_{comp}(\mathbf{R}_+)$ . This gives existence and uniqueness for initial values in the domain  $D(A) \supset (x+1)^{-1}L^1(\mathbf{R}_+)$  of  $A$  if we relax the decay condition  $\int_0^\infty x|P(x,t)|dx < \infty$  for  $t > 0$  to  $P(\cdot, t) \in D(A)$  when  $t > 0$ . The existence and uniqueness as stated above follows from the fact that when  $t > 0$ ,  $e^{-tA}P_0$  is in fact exponentially bounded:

$$\int_0^\infty e^{cx}|(e^{-tA}P_0)(x)|dx < \infty, \quad \text{when } c < 1, c < t \text{ and } t > 0.$$

This bound follows by iteration in Duhamel's principle

$$e^{-tA}P_0 = \int_0^t e^{-2(t-s)}T(t-s)A_c e^{-sA}P_0 ds + e^{-2t}T(t)P_0,$$

together with the following explicit formula for  $T(t)$ ,

$$(T(t)P_0)(x) = e^{-tx}P_0(x) + 2te^{-tx} \int_x^\infty x/yP_0(y)dy + t^2xe^{-tx} \int_x^\infty (y-x)/yP_0(y)dy.$$

Having given a well-posed mathematical formulation of the problem, the main problem is to study the behavior of the solution as  $t \rightarrow \infty$ . For chemical reasons one would expect that  $P(x,t) \rightarrow xe^{-x}$  as  $t \rightarrow \infty$ . In fact, there exist linear functionals  $a_n$  on  $L^1(\mathbf{R}_+)$ , functions  $A_n$  in  $L^1(\mathbf{R}_+)$  and a sequence  $(\mu_n)_{n=2}^\infty$  of positive numbers increasing to 2 as  $n \rightarrow \infty$  such that if  $P_0 \in \mathcal{P}$  and  $N \in \mathbf{N}$ , we have in  $L^1(\mathbf{R}_+)$  as  $t \rightarrow \infty$ ,

$$P(x,t) = xe^{-x} + \sum_{n=2}^N a_n(P_0)A_n(x)e^{-t\mu_n} + O(e^{-t\mu_{N+1}}).$$

The  $A_n(x)$  are, of course, eigenfunctions of  $A$  with eigenvalues  $\mu_n$ . This describes  $P(x,t)$  up to  $O(e^{-2t})$ . The main point of interest for chemists is probably the value of  $\mu_2$ , which gives the rate of approach to equilibrium. By numerical calculations,  $\mu_2$  is close to 1.506. The proof of this asymptotic formula rests on the introduction of an auxilliary selfadjoint operator in  $L^2(\mathbf{R}_+)$ . When  $\psi \in L^2(\mathbf{R}_+)$ , the function  $\sqrt{x}e^{-x/2}\psi(x)$  belongs to  $L^1(\mathbf{R}_+)$  and we define

$$H\psi(x) = (\sqrt{x}e^{-x/2})^{-1}A_1(\sqrt{x}e^{-x/2}\psi(x)) - 2\psi(x).$$

This operator is studied in Section 2 below, we refer to Theorem 1 there. By the spectral theorem, we have if  $\chi_n$  are the normalized eigenfunctions of  $H$  that

$$e^{-t(H+2)}\psi = \sum_{n=1}^\infty (\psi, \chi_n)\chi_n e^{-t(\lambda_n+2)} + \int_0^\infty e^{-t(\lambda+2)}dE(\lambda)\psi.$$

The asymptotic formula follows when  $P_0 = \sqrt{x}e^{-x/2}\psi$  belongs to  $\sqrt{x}e^{-x/2}L^2(\mathbf{R}_+)$  if we let  $a_n(P_0) = (\psi, \chi_n)$ ,  $A_n(x) = \sqrt{x}e^{-x/2}\chi_n(x)$  and  $\mu_n = \lambda_n + 2$ . In general the formula follows from (the proof of) the exponential bound of  $P(\cdot, t)$  mentioned above, since it follows that the distance from  $P(\cdot, t)$  to  $\sqrt{x}e^{-x/2}L^2(\mathbf{R}_+)$  is  $O(e^{-2t})$  as  $t \rightarrow \infty$ . We refer to [Ko1] and [Ko2] for further details.

**2. A selfadjoint operator in  $L^2(\mathbf{R}_+)$ .** When  $H$  is defined as in Section 1, it has the expression

$$H\psi(x) = x\psi(x) + \int_0^\infty v(x,y)\psi(y) dy, \quad v(x,y) = \begin{cases} -2\sqrt{x}e^{x/2}(\sqrt{y}e^{y/2})^{-1}, & 0 < x < y, \\ -2\sqrt{y}e^{y/2}(\sqrt{x}e^{x/2})^{-1}, & 0 < y < x. \end{cases}$$

**THEOREM 1.**  $H$  is selfadjoint on  $\{\varphi \in L^2(\mathbf{R}_+) \mid x\varphi \in L^2(\mathbf{R}_+)\}$ . The spectrum of  $H$  is  $\{\lambda_n\} \cup [0, \infty[$ , where  $[0, \infty[$  is purely absolutely continuous,  $\lambda_1 = -2$  and  $\lambda_n < 0$  increases to 0 as  $n \rightarrow \infty$ . With  $\varepsilon = \exp(-2\pi/\sqrt{7})$ , there exist constants  $a_{kj} \in \mathbf{R}$  for  $j, k \in \mathbf{N}_0$ ,  $j \leq k/2$  with  $a_{00} > 0$  such that as  $n \rightarrow \infty$ ,

$$\lambda_n \sim -\varepsilon^n \sum_{k=0}^\infty \sum_{j=0}^{[k/2]} a_{kj} n^j \varepsilon^{nk/2}.$$

In the rest of this section we describe the proof of this theorem. Note first that  $V\psi(x) = \int_0^\infty v(x,y)\psi(y) dy$  is a relatively compact perturbation of  $x$ . In fact,  $V(x+1)^{-1}$  is a Hilbert-Schmidt operator:

$$\iint |v(x,y)/(y+1)|^2 dx dy \leq 4 \int_0^\infty (y+1)^{-2} dy \int_{-\infty}^\infty e^{-|x-y|} dx = 8.$$

The selfadjointness of  $H$  follows from a theorem of Kato and Rellich; and by a theorem of Weyl, the essential spectrum of  $H$  is equal to the essential spectrum  $[0, \infty[$  of  $x$ . The absence of singular continuous spectrum and discreteness of embedded eigenvalues may be proved by various methods of spectral and scattering theory. One may prove limiting absorption principles, i.e. boundary values at the positive real axis (away from a discrete subset) of the resolvent  $(H - z)^{-1}$  in weighted Sobolev spaces,

$$(H - \lambda \mp i0)^{-1} : \sqrt{\frac{1+x}{x}} \dot{H}^{(s)}(\mathbf{R}_+) \rightarrow \sqrt{\frac{x}{1+x}} \bar{H}^{(-s)}(\mathbf{R}_+), \quad 1/2 < s < 1.$$

(The Sobolev space  $H^{(s)}(\mathbf{R})$  is defined by  $(\int(1+|\xi|^2)^s |\hat{u}(\xi)|^2 d\xi)^{1/2} < \infty$ . The subspace of  $u$  with support in  $[0, \infty[$  is  $\dot{H}^{(s)}(\mathbf{R}_+)$ , and  $\bar{H}^{(-s)}(\mathbf{R}_+)$  is the space of restrictions to  $\mathbf{R}_+$  of elements of  $H^{(-s)}(\mathbf{R}_+)$ ). By theorems of Kato and Kuroda, [K-K], we get absence of singular continuous spectrum, discreteness of embedded eigenvalues and, moreover, existence and asymptotic completeness of the wave operators

$$W_\pm = s\text{-}\lim_{t \rightarrow \pm\infty} e^{itH} e^{-itx}.$$

Asymptotic completeness means that the  $W_\pm$  are partial isometries with initial domain  $\mathcal{H}$  and range  $\mathcal{H}_{ac}(H)$ . Thus the  $W_\pm$  give unitary equivalences of  $x$  and the spectrally continuous part of  $H$ . One may also prove absence of singular continuous spectrum and discreteness of embedded eigenvalues using complex scaling, cf. Aguiar and Combes [A-C], in fact,  $V$  is a dilation analytic perturbation of  $x$ . Finally, existence and asymptotic completeness of the wave operators also follows from a theorem of Kato and Rosenblum, [K,sect. X.4.4], because  $(x+1)^{-1}V(x+1)^{-1}$  is a trace class operator.

The hardest part of the proof of Theorem 1 is the study of the point spectrum. The form of  $v(x, y)$  suggests that it should be the Green's function of a second order Sturm-Liouville problem. We seek an operator  $Lu = (pu')' + qu$  such that  $L(\sqrt{x}e^{x/2}) = 0$  and  $L(\sqrt{x}^{-1}e^{-x/2}) = 0$ . This determines  $(p, q)$  up to a constant factor. The choice

$$Lu = \left(\frac{x}{2x+2}u'\right)' - \frac{x+1}{8x}u,$$

implies by a direct computation that  $LV\varphi = \varphi$  when  $\varphi \in L^2(\mathbf{R}_+)$ . We can now reformulate the eigenvalue equation as an ordinary differential equation. For  $\lambda < 0$  and  $u = (x - \lambda)\varphi$ , we have  $\varphi \in D(H)$  if and only if  $u \in L^2(\mathbf{R}_+)$ ; and when this is true,

$$H\varphi = \lambda\varphi \iff (x - \lambda)\varphi + V\varphi = 0 \iff Lu + (x - \lambda)^{-1}u = 0.$$

The last implication follows from  $LV = 1$  on  $L^2(\mathbf{R}_+)$  and  $Lw = 0 \Rightarrow w = 0$  when  $w \in L^2(\mathbf{R}_+)$ . Thus  $\lambda < 0$  is an eigenvalue if and only if there exists a solution  $u \in L^2(\mathbf{R}_+) \setminus \{0\}$  of the equation

$$\left(\frac{x}{2x+2}u'\right)' - \left(\frac{x+1}{8x} - \frac{1}{x-\lambda}\right)u = 0.$$

This equation has rational coefficients and singularities at 0,  $-1$ ,  $\lambda$  and  $\infty$ . We can compute the behavior of solutions near the singular points using the classical theory of ordinary differential equations, cf. Coddington and Levinson [C-L]. Whether a solution on  $\mathbf{R}_+$  is in  $L^2(\mathbf{R}_+)$  is determined by the asymptotic behavior at 0 and  $\infty$ .

The singularity at  $\infty$  is irregular. There is a basis of solutions on  $\mathbf{R}_+$  consisting of

$$u_\infty(x, \lambda) \sim x^{3/2}e^{-x/2} \quad \text{and} \quad \tilde{u}_\infty(x, \lambda) \sim x^{-3/2}e^{x/2} \quad \text{as } x \rightarrow \infty.$$

This is in fact true for all  $\lambda \in \mathbf{C}$  and  $u_\infty(x, \lambda)$  is an analytic function of  $\lambda$  (only defined on  $] \lambda, \infty[$  if  $\lambda > 0$ ).

When  $\lambda < 0$  the singularity at 0 is regular. There is a basis of solution on  $\mathbf{R}_+$  consisting of

$$u_0(x, \lambda) \sim x^{1/2} \quad \text{and} \quad \tilde{u}_0(x, \lambda) \sim x^{-1/2} \quad \text{as } x \rightarrow 0+.$$

Thus  $\lambda < 0$  is an eigenvalue if and only if  $u_\infty(x, \lambda)$  and  $u_0(x, \lambda)$  are proportional, or equivalently, if and only if  $u_\infty(0+, \lambda) = 0$ .

When  $\lambda = 0$  the singularities at 0 and  $\lambda$  collide, but we still get a regular singularity at 0 this time with a solution basis of the form

$$w(x) = x^{-i\sqrt{7}/2}(1 + O(x)) \quad \text{and} \quad \bar{w}(x) \quad \text{as } x \rightarrow 0+,$$

where  $O(x)$  is analytic in a neighborhood of 0. It follows that  $u_\infty(x, 0)$  is a real valued linear combination of  $w$  and  $\bar{w}$ , and so for some  $\alpha_0 \in \mathbf{C} \setminus \{0\}$ ,

$$u_\infty(x, 0) = \Im(\alpha_0 w(x)) = (\alpha_0' + O(x)) \sin\left(-\frac{\sqrt{7}}{2} \log x + \alpha_0'' + O(x)\right).$$

In particular,  $x^{-1}u_\infty(x, 0)$  is not in  $L^2(\mathbf{R}_+)$ , and so 0 is not an eigenvalue. To prove absence of embedded eigenvalues requires a study of the singular point  $\lambda$  too. We refer to [Ko1] and [Ko2] for this and turn towards the negative eigenvalues.

LEMMA 2. *There are infinitely many negative eigenvalues of  $H$ . The  $n$ 'th eigenfunction has exactly  $n$  zeroes on  $[0, \infty[$ .*

PROOF: Since  $\frac{1}{x-\lambda}$  is an increasing function of  $\lambda < 0$  for fixed  $x > 0$ , it follows from the oscillation theorem of Sturm cf. [C-L,p.208] that the number of zeroes of  $u_\infty(\cdot, \lambda)$  on  $[0, \infty[$  is a non-decreasing function of  $\lambda$ ; and when the  $n$ 'th zero (counted from above) exists, it is an increasing function of  $\lambda$ . Now  $u_\infty(x, -2) = (x+2)\sqrt{x}e^{-x/2}$ , which has no zeroes in  $\mathbf{R}_+$ , so  $-2$  is the lowest eigenvalue. On the other hand,  $u_\infty(x, 0)$  has infinitely many zeroes by the expression above. New zeroes of  $u_\infty(\cdot, \lambda)$  as  $\lambda$  increases must appear at  $x = 0$ , and the corresponding values of  $\lambda$  are eigenvalues. Thus there are infinitely many eigenvalues. Since  $\frac{x+1}{8x} - \frac{1}{x-\lambda} > 0$  when  $0 < x < -\lambda/7$ , there can be at most one zero of  $u_\infty(\cdot, \lambda)$  in  $[0, -\lambda/7]$  for each  $\lambda$ . In particular, the zeroes can not appear in pairs at  $x = 0$  as  $\lambda$  increases, so the number of zeroes of  $u_\infty(\cdot, \lambda_n)$  in  $[0, \infty[$  is exactly  $n$ .

We need a better estimate of the convergence of  $u_\infty(x, \lambda)$  to  $u_\infty(x, 0)$  as  $\lambda \rightarrow 0-$ . The right idea is to use a variation of parameters approach. In principle we rewrite the equation for  $u_\infty(x, \lambda)$  as a first order equation for  $(u_\infty(x, \lambda), \frac{x}{2x+2}u'_\infty(x, \lambda))$  and study the equation for  $W(x)^{-1}$  times this vector, where  $W(x)$  is a fundamental matrix for the equation for  $(u_\infty(x, 0), \frac{x}{2x+2}u'_\infty(x, 0))$ . To exploit the fact that  $u_\infty(x, \lambda)$  is real valued we formulate this approach as follows. Find  $\alpha_\lambda(x)$  such that

$$u_\infty(x, \lambda) = \Im(\alpha_\lambda(x)w(x)), \quad u'_\infty(x, \lambda) = \Im(\alpha_\lambda(x)w'(x)).$$

Then  $\alpha_\lambda(x) = \frac{x}{2x+2}(u'_\infty(x, \lambda)\bar{w}(x) - u_\infty(x, \lambda)\bar{w}'(x))/\Omega$ , where  $\Omega$  is a constant, and we have the equation for  $\alpha_\lambda$ ,

$$\alpha'_\lambda(x) = \Im(\alpha_\lambda(x)w(x))\bar{w}(x)(\frac{1}{x} - \frac{1}{x-\lambda})/\Omega.$$

In particular,  $\alpha_0(x) = \alpha_0 = \text{constant}$ . It follows that when  $0 < x < 1$  and  $\lambda < 0$  we have  $|\alpha'_\lambda(x)| \leq C|\alpha_\lambda(x)||\lambda|/x^2$ . Since  $\int_x^\infty |\lambda|y^{-2} dy = |\lambda|/x$ , we get

$$|\alpha_\lambda(x) - \alpha_0| \leq C'|\lambda|/x \text{ when } |\lambda| < x < 1.$$

Inserting this into  $u_\infty(x, \lambda) = \Im(\alpha_\lambda(x)w(x))$ , we find that if  $T > 0$  is large enough,

$$u_\infty(x, \lambda) = A(x, \lambda) \sin(-\frac{\sqrt{7}}{2} \log x + a_\infty + O(x + |\lambda|/x)), \quad T|\lambda| < x < T^{-1},$$

with  $A(x, \lambda) \neq 0$  there. Thus the zeroes of  $u_\infty(x, \lambda)$  in  $[T|\lambda|, T^{-1}]$  is given by the condition on the phase that  $-\frac{\sqrt{7}}{2} \log x + a_\infty + O(x + |\lambda|/x) \in \pi\mathbf{Z}$ . If we choose  $a_\infty$  suitably, the value of the phase will be  $k\pi$  at the  $k$ 'th zero.

Similarly, we get after a change of variables  $x = -\lambda/\xi$  that

$$u_0(x, \lambda) = B(x, \lambda) \sin(-\frac{\sqrt{7}}{2} \log(-\lambda/x) + b_0 + O(x + |\lambda|/x)), \quad T|\lambda| < x < T^{-1}, \quad \lambda < 0,$$

with  $B(x, \lambda) \neq 0$  there.

Now, consider the  $n$ 'th eigenvalue  $\lambda_n$  for large  $n$ . The phase of  $u_\infty(x, \lambda_n)$  at  $x = \varepsilon^{\pm 1} \sqrt{|\lambda_n|} \in [T|\lambda_n|, T^{-1}]$  is

$$\pm\pi - \frac{\sqrt{7}}{2} \log \sqrt{|\lambda_n|} + a_\infty + O(\sqrt{|\lambda_n|}),$$

and so if  $n$  is large enough, there is a zero  $x_n$  of  $u_\infty(\cdot, \lambda_n)$  in  $[\varepsilon\sqrt{|\lambda_n|}, \varepsilon^{-1}\sqrt{|\lambda_n|}]$ . Since the  $n$ 'th eigenfunction has  $n$  zeroes we get

$$-\frac{\sqrt{7}}{2} \log x_n + a_\infty + O(\sqrt{|\lambda_n|}) - \frac{\sqrt{7}}{2} \log(-\lambda_n/x_n) + b_0 = n\pi.$$

Thus, with  $a_{00} = \exp(2(a_\infty + b_0)/\sqrt{7})$ ,  $\lambda_n = -a_{00}\varepsilon^n + O(\varepsilon^{3n/2})$ , which gives the first term in the asymptotic series of Theorem 1.

To get the full asymptotic series we write the differential equation for  $\alpha_\lambda$  as an integral equation

$$\alpha_\lambda(x) = \alpha_\lambda(x_0) + \int_{x_0}^x \Im(\alpha_\lambda(y)w(y))\bar{w}(y)\left(\frac{1}{y} - \frac{1}{y-\lambda}\right)/\Omega dy.$$

Iterating this equation gives an asymptotic series for  $\alpha_\lambda$  for  $T|\lambda| < x < T^{-1}$ ,

$$\alpha_\lambda(x) \sim \alpha_0 + \sum_{j,k=0}^{\infty} \sum_{l=0}^{k+1} (\lambda/x)^{k+1} x^j (x \log x)^l (b_{jkl} + c_{jkl} x^{i\sqrt{7}}).$$

This implies an asymptotic series for the phase in the expression above for  $u_\infty(x, \lambda)$  and we have a similar asymptotic series for the phase in the expression for  $u_0(x, \lambda)$ . The full asymptotic series for  $\lambda_n$  in Theorem 1 is now proved by an iteration scheme, in which one subsequently finds more and more terms in this series and better and better asymptotic formulas for the zero  $x_n$ . The details may be found in [Ko1] and [Ko2].

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