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On the recovery and resolution of exponential relaxation rates from experimental data: a singular-value analysis of the Laplace transform inversion in the presence of noise

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The problem of numerical inversion of the Laplace transform is considered when the inverse function is of bounded, strictly positive support. The recent eigenvalue analysis of McWhirter and Pike for infinite support has been generalized by numerical calculations of singular values. *A priori* knowledge of the support is shown to lead to increased resolution in the inversion, and the number of exponentials that can be recovered in given levels of noise is calculated.

1. Introduction

In much of experimental science, data delivered by an instrumental system are related to the natural phenomenon under investigation by a 'resolution' limit, which may be expressed by a linear integral transformation K of the form

$$g(p) = (Kf)(p) = \int_F T(p,t)f(t)\,\mathrm{d}t, \quad p \in G. \tag{1.1}$$

This is a Fredholm equation of the first kind: $T(p,t)$ is the kernel of the transformation and defines the effect of the instrument on the 'natural' input $f(t)$ in producing the measured data $g(p)$; F and G are the domains of support of the variable t explored and the variable p measured; they may be multidimensional.

For band-limited imaging or communication systems, $T(p,t)$ has a Bessel-function form, and the analysis of such systems has given rise to a well developed theory of 'resolution' or 'information' associated with the names, for example, of Nyquist, Shannon, Slepian and Pollak, Frieden, and Toraldo di Francia, but dating back to Abbé and Lord Rayleigh. One finds that, in the presence of noise, the 'object' $f(t)$ can only be recovered from the 'image' $g(p)$ up to a limit of resolution (the Shannon or Nyquist number, or Rayleigh criterion), determined by the properties of the eigenvalue spectrum of the transformation $f \to g$, and that finer detail is irrecoverable owing to the 'ill-conditioned' nature of the inversion. This 'classical' theory of information is concerned with the problem of recovering and resolving natural spatial or temporal *oscillatory* components. An important alternative situation in which the experimenter is concerned to recover and resolve *exponential relaxation* rates is described by the same equation with a different kernel. This Fredholm equation has the Laplace-transform kernel

$$T(p,t) = e^{-pt}. \tag{1.2}$$

For the same basic reasons, which we shall take up later, this inversion is also ill conditioned (Bellmann *et al.* 1966; Schoenberg 1973; McWhirter & Pike 1978) but, curiously, has not received the same detailed consideration from an 'information theory' point of view although there is no lack of *ad hoc* attempts in the literature on numerical inversion.

A step towards generalization of the concepts of resolution and information for the recovery of exponentials was taken in a recent paper by McWhirter & Pike (1978) in which they calculated analytically the eigenfunctions and eigenvalues of the Laplace transform and, as a consequence, were able to identify resolution elements and an analogue of the Shannon number for this problem. In contrast to the Shannon number of the previous information theory this new 'Shannon' number is strongly dependent on the experimental noise present. It, nevertheless, determines the maximum number of exponential relaxation rates that may be successfully determined and, in fact, requires that these be spaced in a geometrical sequence in the independent variable.

In a further recent contribution to the classical information-theory problem Bertero & Pike (1982) have shown that significantly improved resolution can be achieved by restriction of the object support with respect to that of the image. The eigenfunction analysis of the extant theory is not appropriate for this situation and a new 'singular-value' analysis of information and resolution was presented. This new theory gives the prospect of real gains in the performance of linear systems, for example the optical microscope, by suitable design.

In a similar vein, the recovery and resolution of exponential object-components envisaged by McWhirter & Pike may be improved upon by using *a priori* knowledge of the support of the object. The present paper is concerned with proving and quantifying this proposition.

The content of the paper is rather mathematical and we therefore explain here the results that will be easily assimilated by the interested reader. These are contained in figure 1. If one has knowledge of the lower and upper bounds $\{a, b\}$ of the support of $f(t)$, then we show that exponential components may be recovered at values of t_0, $\delta_s t_0$, $\delta_s^2 t_0$, $\delta_s^3 t_0$, ... within this support where the 'resolution ratio', δ_s, is given in the figure as a function of the ratio, γ, of b to a for various 'signal:noise ratios', E/ϵ.

In the next section we make some general remarks about the Laplace transform and demonstrate the nature of its ill conditioning.

In § 3 we discuss singular values and singular functions of the Laplace transform and we derive some properties of the singular values like non-degeneracy and asymptotic distribution when the parameter $\gamma = b/a$ tends to infinity. We give also the results of some numerical computations.

In § 4 we consider a very simple noise model and a truncated singular-function expansion for the approximate solution to discuss the resolution limits achievable in the Laplace-transform inversion. Typically we find significant improvements in resolution as the parameter $\gamma = b/a$ approaches unity. The paper is completed by two mathematical Appendixes.

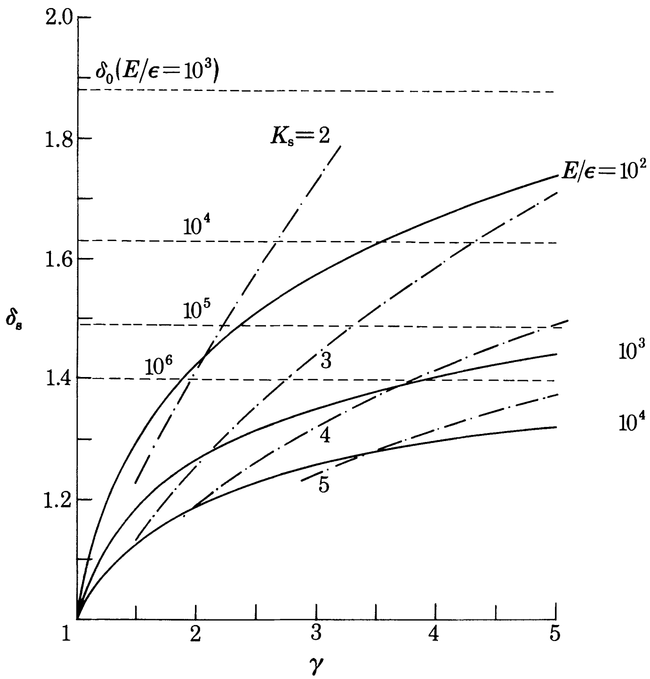

FIGURE 1. The resolution ratio δ_s as a function of the parameter γ and of the signal:noise ratio E/ϵ (see equation (4.21)). The horizontal dashed lines are the values for infinite support. The dash–dot lines give the number of exponentials that may be recovered ('Shannon' number).

In a future publication we shall consider the effects of the necessarily finite support of the data and give optimum placings for the upper and lower observation limits for given fixed numbers of data points. We shall also publish separately a detailed application of the method to the analysis of macromolecular diffusion by light scattering.

2. The Laplace transform

Inserting the kernel (1.2) into equation (1.1) gives explicitly the Laplace transform: for (F, G) equal to $(0, +\infty)$

$$g(p) = \int_0^{+\infty} \mathrm{e}^{-pt} f(t)\,\mathrm{d}t, \quad 0 \leqslant p < +\infty. \tag{2.1}$$

We can define the direct problem as a linear mapping $f \to g$, which is continuous and injective in $L^2(0, +\infty)$. The continuity of the mapping follows from the inequality

$$\int_0^{+\infty} |g(p)|^2 \,\mathrm{d}p \leqslant \pi \int_0^{+\infty} |f(t)|^2 \,\mathrm{d}t, \tag{2.2}$$

which can be derived by applying a Mellin transformation to equation (2.1). Indeed, with the Mellin transform $\tilde{f}$ of a function $f \in L^2(0, +\infty)$ as defined by Titchmarsh (1948),

$$\mathscr{F}(\tfrac{1}{2}+\mathrm{i}\omega) = \tilde{f}(\omega) = \int_0^{+\infty} f(t)\, t^{-\frac{1}{2}+\mathrm{i}\omega} \,\mathrm{d}t, \tag{2.3}$$

from equation (2.1) we get

$$\tilde{g}(\omega) = \Gamma(\tfrac{1}{2}+\mathrm{i}\omega)\tilde{f}(-\omega); \tag{2.4}$$

then inequality (2.2) follows from the Parseval relation (Titchmarsh 1948)

$$\int_{-\infty}^{+\infty} |f(t)|^2 \,\mathrm{d}t = \frac{1}{2\pi}\int_{-\infty}^{+\infty} |\tilde{f}(\omega)|^2 \,\mathrm{d}\omega$$

and from the inequality $|\Gamma(\frac{1}{2}+\mathrm{i}\omega)|^2 = \pi/\cosh \pi\omega \leqslant \pi$.

The inverse mapping $g \to f$, however, is not continuous. An elementary proof of this fact is as follows. Let $\{a_n\}$, $\{b_n\}$ $(n = 1, 2, \ldots)$ be sequences of positive numbers such that a_n, $b_n \to +\infty$, when $n \to +\infty$; then let us consider the following sequence of functions:

$$f_n(t) = (2b_n/\pi)^{\frac{1}{2}} \exp\left[-(1+\mathrm{i}a_n)\,t\right] \tag{2.6}$$

with the associated Laplace transforms

$$g_n(p) = (2b_n/\pi)^{\frac{1}{2}}\,(p+1+\mathrm{i}a_n)^{-1}. \tag{2.7}$$

Very simple computations show that

$$\|f_n\|^2 = \int_0^{+\infty} |f_n(t)|^2 \,\mathrm{d}t = (1/\pi)\,b_n, \tag{2.8}$$

$$\|g_n\|^2 = \int_0^{+\infty} |g_n(p)|^2 \,\mathrm{d}p = \frac{b_n}{a_n}\left[1-\frac{2}{\pi}\arctan\left(\frac{1}{a_n}\right)\right]. \tag{2.9}$$

Therefore, if we choose a_n, b_n in such a way that $b_n/a_n \to 0$ as $n \to +\infty$ (for instance $a_n = b_n^2$), we get $\|f_n\| \to +\infty$, while $\|g_n\| \to 0$, as $n \to +\infty$.

The previous remark implies that, when we know only an approximation g of the exact Laplace transform $\bar{g}$, the solution of the problem is completely indeterminate: the set of functions whose Laplace transforms approximate $\bar{g}$ within a given error is not bounded with respect to the norm of $L^2(0, +\infty)$.

To reduce the uncertainty in the solution one needs further constraints, as are required in regularization theory (Tikhonov & Arsenin 1977; Miller 1970; Bertero *et al.* 1980). However, it must be pointed out that the problem of the numerical inversion of the Laplace transform is severely ill posed. This statement can be justified as follows.

Let $\bar{f}$ be the exact solution corresponding to the exact Laplace transform $\bar{g}$ and let $f \in L^2(0, +\infty)$ be any function whose Laplace transform g approximates $\bar{g}$ in the sense that the L^2-norm of $v = g - \bar{g}$ does not exceed a given positive number ϵ. If we now write $u = f - \bar{f}$ (v is the Laplace transform of u), then from equation (2.4) and from the Parseval relation (2.5) we get

$$\int_0^{+\infty} |v(p)|^2 \,\mathrm{d}p = \frac{1}{2\pi}\int_{-\infty}^{+\infty} \frac{\pi}{\cosh \pi\omega} |\tilde{u}(\omega)|^2 \,\mathrm{d}\omega \leqslant \epsilon^2. \tag{2.10}$$

Let us also assume a constraint on the first derivative of u:

$$\int_0^{+\infty} t|u'(t)|^2 \,\mathrm{d}t = \frac{1}{2\pi}\int_{-\infty}^{+\infty} (\omega^2 + \tfrac{1}{4})\, |\tilde{u}(\omega)|^2 \,\mathrm{d}\omega \leqslant E^2. \tag{2.11}$$

Now let $S \subset L^2(0, +\infty)$ be the subset of the functions u satisfying the constraints (2.10), (2.11) and let S_0 be the subset of the functions u satisfying the constraint

$$\frac{1}{2\pi}\int_{-\infty}^{+\infty} \left[\frac{\pi}{\cosh \pi\omega} + \left(\frac{\epsilon}{E}\right)^2 (\omega^2 + \tfrac{1}{4})\right] |\tilde{u}(\omega)|^2 \,\mathrm{d}\omega \leqslant \epsilon^2. \tag{2.12}$$

Clearly $S_0 \subset S$ and therefore the diameter of S_0 gives a lower bound for the uncertainty in the functions f, satisfying the constraint (2.11), whose Laplace transforms g approximate $\bar{g}$ within the error ϵ. In Appendix A it is proved that, for small ϵ/E,

$$M_0(\epsilon, E) = \sup_{u \in S_0} \|u\| \sim \pi E/2|\ln(\epsilon/E)|. \tag{2.13}$$

As a consequence the uncertainty in the solution tends to zero very slowly when the error in the data tends to zero. The uncertainty is considerably reduced, however, when much more restrictive smoothness conditions are satisfied. For instance, let f admit an analytic continuation $f[t\exp(\mathrm{i}\phi)]$ in the angular sector $|\phi| < \alpha$ and let $f[t\exp(\mathrm{i}\alpha)]$ be square integrable. These conditions are satisfied, for instance, by $f(t) = P(t)\exp(-t)$, where $P(t)$ is a polynomial, for any $\alpha = \frac{1}{2}\pi - \eta$ ($\eta > 0$, arbitrary). Now, it is quite easy to prove that the Mellin transform of a function f satisfying the previous conditions has the following asymptotic behaviour:

$$|\tilde{f}(\omega)| \sim C\exp(-\alpha|\omega|), \quad |\omega| \to +\infty.$$

As a consequence we can assume that the function $u = f - \bar{f}$ satisfies the constraint

$$\frac{1}{2\pi}\int_{-\infty}^{+\infty} \cosh(2\alpha\omega)\, |\tilde{u}(\omega)|^2 \,\mathrm{d}\omega \leqslant E^2. \tag{2.14}$$

We can combine again the two constraints (2.10) and (2.14) into the single one

$$\frac{1}{2\pi}\int_{-\infty}^{+\infty} \left[\frac{\pi}{\cosh(\pi\omega)} + \left(\frac{\epsilon}{E}\right)^2 \cosh(2\alpha\omega)\right] |\tilde{u}(\omega)|^2 \,\mathrm{d}\omega \leqslant \epsilon^2 \tag{2.15}$$

and denote by S_0 the subset of the functions u satisfying (2.15). Then in Appendix A it is proved that, for small ϵ/E,

$$M_0(\epsilon, E) = \sup_{u \in S_0} \|u\| \sim \text{const.} \times E\left(\frac{\epsilon}{E}\right)^{\beta}, \quad \beta = \frac{2\alpha}{\pi + 2\alpha}. \tag{2.16}$$

We see that, in this case, the uncertainty in the solution tends to zero rapidly and therefore accurate results can be obtained in the Laplace-transform inversion, by using either eigenfunction expansions (McWhirter & Pike 1978) or any other method, for instance best-rational-function approximations (Longman 1974).

Unfortunately, the condition of analyticity of the solution is not satisfied in some important applications of the Laplace-transform inversion. An important example is the analysis of light scattering polydispersity (Ostrowski *et al.* 1981), where it is known that the function f has a bounded support and therefore it cannot be analytic; on the other hand it is quite obvious that, if one takes into account the knowledge of the support of f, one has to get a reduction of the uncertainty in the solution with respect to the situation described by equations (2.12), (2.13).

3. Singular values and singular functions of the Laplace transform

The starting point of our investigation is the following remark: if we consider functions f supported in the interval $[a, b]$, $a > 0$, then the linear mapping $f \rightarrow g$ defines a compact, injective operator of $L^2(a, b)$ into $L^2(0, +\infty)$. As a consequence, the well known singular-value method for the solution of Fredholm integral equations of the first kind (Miller 1974) can be applied to the Laplace-transform inversion.

Let us consider the restriction of (2.1) to the class of square integrable functions supported in the interval $[a, b]$, $a > 0$; in such a case the Laplace transform $g(p)$ is an entire analytic function and, for real p, $g(p)$ tends to zero exponentially fast when $p \rightarrow +\infty$.

The mapping $f \rightarrow g$ defines the following linear operator from $L^2(a, b)$ into $L^2(0, +\infty)$:

$$(Kf)(p) = \int_a^b e^{-pt} f(t)\, dt, \quad 0 \leqslant p < +\infty. \tag{3.1}$$

As follows from equation (2.2), K is a continuous operator; it is also quite easy to prove that K is compact. Indeed the image of a bounded set of $L^2(a, b)$ under the operator K is a set of equicontinuous functions, uniformly bounded by an exponential function. Also, from the well known properties of the Laplace transform it follows that the equation $Kf = 0$ has only the trivial solution $f = 0$ and therefore the operator K is injective.

The adjoint operator K^* is given by

$$(K^*g)(t) = \int_0^{+\infty} e^{-tp}\, g(p)\, dp, \quad a \leqslant t \leqslant b; \tag{3.2}$$

it is a linear, compact operator from $L^2(0, +\infty)$ into $L^2(a, b)$. Since K is injective, the range of K^* is dense in $L^2(a, b)$; K^* is also injective and therefore the range of K is dense in $L^2(0, +\infty)$.

From the previous properties it follows that the operator K admits a singular system $\{\alpha_k; u_k, v_k\}$ ($k = 0, 1, \ldots$), given by the solutions of the coupled equations

$$Ku_k = \alpha_k v_k, \quad K^* v_k = \alpha_k u_k. \tag{3.3}$$

Since the null spaces of K and K^* contain only the null element (of $L^2(a, b)$ and $L^2(0, +\infty)$ respectively) we reach the conclusion that all the singular values α_k are strictly positive, that the set $\{u_k\}$ ($k = 0, 1, 2, \ldots$) is a basis in $L^2(a, b)$ and that the set $\{v_k\}$ ($k = 0, 1, 2, \ldots$) is a basis in $L^2(0, +\infty)$.

As is well known, the singular functions u_k are the eigenfunctions of the operator K^*K associated with the eigenvalues α_k^2:

$$K^*K\, u_k = \alpha_k^2 u_k, \quad k = 0, 1, 2, \ldots; \tag{3.4}$$

as usual we assume that the eigenvalues α_k^2 are ordered in a non-increasing sequence $\alpha_0^2 \geqslant \alpha_1^2 \geqslant \alpha_2^2 \geqslant \ldots$.

From equations (3.1), (3.2) it is easy to derive that

$$(K^*Kf)(t) = \int_a^b \frac{f(s)}{t+s}\,\mathrm{d}s, \quad a \leqslant t \leqslant b, \tag{3.5}$$

and therefore K^*K is an operator of the trace class:

$$\operatorname{tr}(K^*K) = \sum_{k=0}^{+\infty} \alpha_k^2 = \int_a^b \frac{\mathrm{d}t}{2t} = \tfrac{1}{2}\ln\gamma, \tag{3.6}$$

where

$$\gamma = b/a. \tag{3.7}$$

It is important to remark that, as suggested by equation (3.6), the singular values α_k of the operator (3.1) depend only on the parameter γ. Indeed, writing explicitly the eigenvalue equation (3.4),

$$\int_a^b \frac{u_k(s)}{t+s}\,\mathrm{d}s = \alpha_k^2 u_k(t), \tag{3.8}$$

and introducing the new variables

$$t = a + (b-a)x, \quad s = a + (b-a)y \tag{3.9}$$

and the parameter

$$\beta = 2a/(b-a) = 2/(\gamma - 1) \quad (\gamma = 1 + 2/\beta), \tag{3.10}$$

we obtain the following eigenvalue problem:

$$\int_0^1 \frac{\psi_k(y)}{x+y+\beta}\,\mathrm{d}y = \alpha_k^2 \psi_k(x). \tag{3.11}$$

The eigenvalues α_k^2 are the same in equations (3.8) and (3.11) and the normalized eigenfunctions are related by

$$u_k(t) = \frac{1}{(b-a)^{\frac{1}{2}}}\psi_k\left(\frac{t-a}{b-a}\right). \tag{3.12}$$

More precise properties of the eigenvalues of the operator (3.5) are derived in Appendix B, with the use of results concerning the spectrum of Toeplitz operators (Kac *et al.* 1953; Landau 1975; Gori & Palma 1975). In this way we show that

(i) each eigenvalue of K^*K is non-degenerate, i.e. the corresponding eigenspace is one-dimensional;

(ii) if $N_\gamma(\delta_0, \delta_1)$ is the number of eigenvalues of K^*K satisfying the condition $\delta_0 < \alpha_k^2 < \delta_1 (\delta_0 > 0)$ and if $\mu^+(\delta_0, \delta_1)$ is the measure of the ω-interval, $\omega > 0$, where

$$\delta_0 < \pi/\cosh \pi\omega < \delta_1, \tag{3.13}$$

then

$$\lim_{\gamma\to\infty} \frac{N_\gamma(\delta_0, \delta_1)}{\ln\gamma} = \frac{1}{\pi}\mu^+(\delta_0, \delta_1). \tag{3.14}$$

Property (ii) implies that the eigenvalues of K^*K, when $\gamma \to +\infty$, approach the Mellin transform of the kernel $K(t) = (1+t)^{-1}$ in a well defined sense (remark that in the limit $\gamma = +\infty$, which corresponds either to $a = 0$ or to $b = +\infty$, there is a change in the nature of the spectrum of K^*K; the spectrum becomes continuous and coincides with the interval $[0, \pi]$). Indeed, when γ is very large, equation (3.14) gives an approximate formula for $N_\gamma(\delta_0, \delta_1)$, which implies the following result: if only one eigenvalue falls in the interval (δ_0, δ_1), then $\mu^+(\delta_0, \delta_1) \sim \pi/\ln\gamma$ (this quantity is just one-half the Nyquist distance for the Mellin transform of a function supported in $[a, b]$). As a consequence we get the following approximate formula for the eigenvalues of K^*K:

$$\alpha_k^2 \sim \pi/\cosh \pi\omega_k, \quad \omega_k = (\pi/\ln\gamma)\, k. \tag{3.15}$$

In particular this equation implies that the greatest singular value α_0 of K tends to $\pi^{\frac{1}{2}}$ when $\gamma \to +\infty$. This result is in agreement with the following inequality:

$$\alpha_0 \leqslant \min\{\pi^{\frac{1}{2}}, \beta^{-\frac{1}{2}}\} \tag{3.16}$$

where β is the parameter defined in equation (3.9). Therefore α_0 is always smaller than $\pi^{\frac{1}{2}}$ and it tends to $\pi^{\frac{1}{2}}$ when $\gamma \to +\infty$ $(\beta \to 0)$; on the other hand α_0 tends to zero at least as $\beta^{-\frac{1}{2}}$ when $\beta \to +\infty$ $(\gamma \to 1)$.

The inequality (3.16) can be proved as follows. From the inequality (2.2), applied to the case where f is supported in $[a, b]$, we derive $\|Kf\| \leqslant \pi^2 \|f\|$. Therefore, taking $f = u_0$ and using equation (3.3) and the normalization property of u_0, v_0, we get $\alpha_0 \leqslant \pi^{\frac{1}{2}}$. On the other hand, from equation (3.5) and the Schwarz inequality, we get

$$\begin{aligned} \|K^*Kf\| &= \left[\int_a^b \left|\int_a^b \frac{f(s)}{t+s}\,\mathrm{d}s\right|^2 \mathrm{d}t\right]^{\frac{1}{2}} \\ &\leqslant \frac{(b-a)^{\frac{1}{2}}}{2a}\int_a^b |f(s)|\,\mathrm{d}s \leqslant \frac{1}{\beta}\|f\| \end{aligned} \tag{3.17}$$

so that, by taking $f = u_0$, we get $\alpha_0^2 \leqslant 1/\beta$.

We have computed numerically the eigenvalues of equation (3.11), approximating the kernel by tensor products of splines (Hämmerlin & Schumaker 1980). In table 1 we report the singular values α_k for various values of the parameter β. The corresponding values of γ are 5, 3, 2, $\frac{5}{3}$, $\frac{3}{2}$, $\frac{7}{5}$, $\frac{4}{3}$ and $\frac{9}{7}$.

TABLE 1

	$\beta = 0.5$	$\beta = 1$	$\beta = 2$	$\beta = 3$
α_0	8.751×10^{-1}	7.323×10^{-1}	5.858×10^{-1}	5.040×10^{-1}
α_1	1.935×10^{-1}	1.133×10^{-1}	5.804×10^{-2}	3.696×10^{-2}
α_2	3.827×10^{-2}	1.569×10^{-2}	5.142×10^{-3}	2.424×10^{-3}
α_3	7.434×10^{-3}	2.134×10^{-3}	4.475×10^{-4}	1.562×10^{-4}
α_4	1.435×10^{-3}	2.883×10^{-4}	3.869×10^{-5}	9.997×10^{-6}
α_5	2.765×10^{-4}	3.885×10^{-5}	3.336×10^{-6}	6.380×10^{-7}
α_6	5.325×10^{-5}	5.227×10^{-6}	2.872×10^{-7}	—
α_7	1.029×10^{-5}	7.029×10^{-7}	—	—
α_8	2.006×10^{-6}	—	—	—
α_9	4.007×10^{-7}	—	—	—

	$\beta = 4$	$\beta = 5$	$\beta = 6$	$\beta = 7$
α_0	4.495×10^{-1}	4.097×10^{-1}	3.789×10^{-1}	3.542×10^{-1}
α_1	2.622×10^{-2}	1.985×10^{-2}	1.571×10^{-2}	1.283×10^{-2}
α_2	1.368×10^{-3}	8.602×10^{-4}	5.824×10^{-4}	4.158×10^{-4}
α_3	7.006×10^{-5}	3.661×10^{-5}	2.120×10^{-5}	1.323×10^{-5}
α_4	3.567×10^{-6}	1.548×10^{-6}	7.673×10^{-7}	4.183×10^{-7}
α_5	1.811×10^{-7}	6.537×10^{-8}	—	—

4. Resolution limits in the Laplace-transform inversion

According to general results on equations of the first kind with compact operators (Miller 1974), the solution of the equation $K\bar{f} = \bar{g}$, when it exists, is given by

$$\bar{f}(t) = \sum_{k=0}^{+\infty} \frac{\bar{g}_k}{\alpha_k} u_k(t), \tag{4.1}$$

where

$$\bar{g}_k = \int_0^{+\infty} \bar{g}(p)\, v_k(p)\, \mathrm{d}p \tag{4.2}$$

and $\{\alpha_k;\, u_k, v_k\}$ $(k = 0, 1, 2, \ldots)$ is the singular system of the operator K, (equation (3.1)). The series (4.1) is convergent if and only if

$$\sum_{k=0}^{+\infty} \frac{|\bar{g}_k|^2}{\alpha_k^2} < +\infty, \tag{4.3}$$

i.e. $\bar{g}$ is the Laplace transform of a function supported in $[a, b]$ if and only if it satisfies condition (4.3). In general this condition is not satisfied when $\bar{g}$ is corrupted by noise or experimental errors and the solution of the problem of Laplace-transform inversion does not then exist. One can look for approximate solutions taking into account properties both of the noise and of the unknown solution.

Let us consider for simplicity the Laplace transform corrupted by additive, zero-mean, white noise, i.e. the experimental Laplace transform is given by

$$g(p) = \bar{g}(p) + n(p) = (K\bar{f})(p) + n(p), \tag{4.4}$$

where $\bar{g}$ is the exact Laplace transform associated with $\bar{f}$ and

$$\langle n(p)\, n(p')\rangle = \epsilon^2 \delta(p - p'). \tag{4.5}$$

Let us further assume that the unknown object $\bar{f}$ is also from a zero-mean, white-noise process with power spectrum E^2,

$$\langle \bar{f}(t)\bar{f}(t')\rangle = E^2\delta(t-t'), \tag{4.6}$$

and that the processes $n, \bar{f}$ are uncorrelated. Since $\{v_k\}$ ($k = 0, 1, 2, \ldots$) is a basis in $L^2(0, +\infty)$ and $\{u_k\}$ ($k = 0, 1, 2, \ldots$) is a basis in $L^2(a, b)$, we can write

$$n(p) = \sum_{k=0}^{+\infty} b_k v_k(p), \quad b_k = \int_0^{+\infty} n(p)\, v_k(p)\, \mathrm{d}p, \tag{4.7}$$

$$\bar{f}(t) = \sum_{k=0}^{+\infty} \bar{a}_k u_k(t), \quad \bar{a}_k = \int_a^b \bar{f}(t)\, u_k(t)\, \mathrm{d}t; \tag{4.8}$$

then from equations (4.5), (4.6) we get

$$\langle b_k b_j\rangle = \epsilon^2\delta_{kj}, \tag{4.9}$$

$$\langle \bar{a}_k \bar{a}_j\rangle = E^2\delta_{kj}. \tag{4.10}$$

Since we have assumed the processes $n, \bar{f}$ to be uncorrelated, we have also

$$\langle \bar{a}_k b_j\rangle = 0. \tag{4.11}$$

Now the components of the reconstructed solution are given by

$$a_k = \frac{1}{\alpha_k}\int_0^{+\infty} g(p)\, v_k(p)\, \mathrm{d}p = \bar{a}_k + \frac{b_k}{\alpha_k}, \tag{4.12}$$

and from equations (4.9)–(4.11) we have

$$\langle a_k a_j\rangle = (E^2 + \epsilon^2/\alpha_k^2)\, \delta_{kj}. \tag{4.13}$$

As a consequence, in the inversion procedure we can estimate only those components such that the variance E^2 of the signal $\bar{a}_k$ is greater than the variance ϵ^2/α_k^2 of the noise contribution b_k/α_k, i.e. those components such that

$$\alpha_k \geqslant \epsilon/E. \tag{4.14}$$

Let us assume now that, for a given value of the parameter γ – equation (3.7) – and for a given value of the signal:noise ratio E/ϵ, equation (4.14) is satisfied for $k = 0, 1, \ldots, K_s$. Since it can be shown by numerical computation that the number of zeros of $u_k(t)$ in the interval $[a, b]$ is just equal to k (we believe that this is a general property of the eigenfunctions of Toeplitz and related operators – see Appendix B – when the Fourier or Mellin transform of the kernel is a non-increasing function for $\omega > 0$), we can conclude that in the reconstruction procedure, founded on singular-function expansions, we can recover $M_s = K_s + 1$ 'resolution elements'. The criterion (4.14), however, implies an uncertainty of *ca.* 70% in the $(K_s + 1)$th component; this, practically, reduces to *ca.* 10% in the K_sth component and we therefore, arbitrarily, define the number of resolvable exponentials by K_s.

We assume now that the zeros of $u_k(t)$ are approximately equidistant in the variable $x = \ln[t/(ab)^{\frac{1}{2}}]$, as suggested by the eigenvalue distribution derived in Appendix B and by the exponential sampling methods developed for the analysis of light scattering polydispersity (Ostrowski *et al.* 1981). In other words we assume that the relevant eigenfunctions $u_k(t)$ of the operator (3.5) are approximately given by trigonometric functions in the variable $x = \ln[t/(ab)^{\frac{1}{2}}]$. As a consequence we take as separation points between adjacent resolution elements those given by $t_m = a\delta_s^m$, $m = 0, 1, \ldots, K_s$. Taking the last point equal to b, we get for the resolution ratio δ_s the following formula:

$$\delta_s = \gamma^{1/K_s}, \tag{4.15}$$

γ being defined in equation (3.7).

We have computed K_s and δ_s from the singular values of the operator K, using equation (4.14) and a linear interpolation between adjacent singular values to get a smooth behaviour of the parameters K_s, δ_s as functions of γ and of E/ϵ. Some results are shown in table 2 and a graphical representation is given in figure 1.

Within these theoretically possible resolution limits the problem is well conditioned and inversion may be accomplished by linear least squares fitting.

TABLE 2

	$\beta = 0.5, \gamma = 5$		$\beta = 1, \gamma = 3$		$\beta = 2, \gamma = 2$		$\beta = 3, \gamma = \frac{5}{3}$	
E/ϵ	K_s	δ_s	K_s	δ_s	K_s	δ_s	K_s	δ_s
10^2	2.917	1.736	2.420	1.574	1.908	1.438	1.780	1.332
10^3	4.375	1.445	3.614	1.355	2.882	1.272	2.628	1.214
10^4	5.791	1.320	4.755	1.260	3.850	1.197	3.384	1.163

	$\beta = 4, \gamma = \frac{3}{2}$		$\beta = 5, \gamma = \frac{7}{5}$		$\beta = 6, \gamma = \frac{4}{3}$		$\beta = 7, \gamma = \frac{9}{7}$	
E/ϵ	K_s	δ_s	K_s	δ_s	K_s	δ_s	K_s	δ_s
10^2	1.653	1.278	1.519	1.248	1.377	1.232	1.228	1.227
10^3	2.283	1.194	1.993	1.184	1.972	1.157	1.953	1.137
10^4	2.977	1.145	2.923	1.122	2.860	1.106	2.784	1.094

We remark that we have a lowering in δ_s – and therefore an improvement in resolution – when we have a lowering of γ or an increase of the signal:noise ratio E/ϵ. It is clear that these results give a quantitative answer to the question of the effect on resolution due to the knowledge of the support of f. In particular in figure 1 we have indicated values of δ_s corresponding to $\beta = 0$ ($\gamma = +\infty$). The values have been computed as follows (McWhirter & Pike 1978; McWhirter 1980).

If we have no knowledge of the support of $\bar{f}$, then we can use the Mellin transform for solving the problem of Laplace-transform inversion; see equation (2.4). In such a case, equation (4.14) is replaced by

$$\pi/\cosh \pi\omega \geqslant (\epsilon/E)^2, \tag{4.16}$$

i.e. we can restore only those Mellin components of $\bar{f}$ such that $|\omega| \leqslant \omega_0$, where ω_0 is the unique positive solution of the equation

$$\cosh \pi\omega_0 = \pi(E/\epsilon)^2. \tag{4.17}$$

Now the zeros of the real and the imaginary parts of $\exp(i\omega_0 \ln t)$ satisfy the relation $t_m = t_{m-1} \exp(\pi/\omega_0)$ and therefore we must take

$$\delta_0 = \delta_s(\beta = 0) = \exp(\pi/\omega_0). \tag{4.18}$$

With $E/\epsilon = 10^2, 10^3, 10^4, 10^5, 10^6$ we get, from equation (4.17), $\omega_0 = 3.52, 4.98, 6.45, 7.91, 9.38$ and, from equation (4.18), $\delta_0 = 2.44, 1.88, 1.63, 1.49, 1.40$, respectively.

To relate the signal:noise ratio E/ϵ used in this theory to sampled data, where a mean square error, $\eta = [\langle\eta(p_i)^2\rangle]^{\frac{1}{2}}$, is known at a set of sample points, p_i, of the data and the mean square value of the object or, equivalently, $g(0)$ is normalized to unity, we use the following two relations derived from equations (4.5), (4.6):

$$\eta^2 = \left\langle \left| \frac{1}{d} \int_{p_i}^{p_i+d} n(p)\,\mathrm{d}p \right|^2 \right\rangle = \frac{\epsilon^2}{d}, \tag{4.19}$$

$$1 = \left\langle \left| \int_1^{\gamma} f(t)\,\mathrm{d}t \right|^2 \right\rangle = (\gamma-1)\,E^2 \tag{4.20}$$

where d is the distance, assumed constant, between data points over which the noise is integrated. These may be combined to obtain

$$E/\epsilon = 1/\eta[d(\gamma-1)]^{\frac{1}{2}}. \tag{4.21}$$

The optimum placing of data points, where these may be chosen for the problem, will be discussed in a future contribution. We find that if 100 linearly spaced data points are optimally placed for $1.5 < \gamma < 10$, then $d \sim 0.1$. Geometric spacing of data points is also under investigation (Pike *et al.* 1982).

Appendix A

In this Appendix we prove equations (2.13) and (2.16) of the text. We need the following lemma.

Lemma. *If $S_0 \subset L^2(0, +\infty)$ is the set of the functions u whose Mellin transform $\tilde{u}(\omega)$ satisfies the condition*

$$\frac{1}{2\pi}\int_{-\infty}^{+\infty} \psi(\omega)\,|\tilde{u}(\omega)|^2\,\mathrm{d}\omega \leqslant \epsilon^2, \tag{A 1}$$

where ψ is a continuous, positive function such that $\psi(\omega) \to +\infty$, $|\omega| \to +\infty$, then

$$\sup_{u \in S_0} \|u\| = \epsilon/\psi_0^{\frac{1}{2}}, \tag{A 2}$$

where ψ_0 is the minimum of ψ.

Proof. Since ψ must have a minimum $\psi_0 > 0$, from condition (A 1) we get

$$\psi_0\,\|u\|^2 = \frac{\psi_0}{2\pi}\int_{-\infty}^{+\infty} |\tilde{u}(\omega)|^2\,\mathrm{d}\omega \leqslant \frac{1}{2\pi}\int_{-\infty}^{+\infty} \psi(\omega)\,|\tilde{u}(\omega)|^2\,\mathrm{d}\omega \leqslant \epsilon^2 \tag{A 3}$$

so that $\|u\| \leqslant \epsilon\psi_0^{-\frac{1}{2}}$. Therefore, to prove equation (A 2) it is enough to show that there exists a sequence $\{u_n\} \subset S_0$ such that $\|u_n\|$ tends to $\epsilon\psi_0^{-\frac{1}{2}}$ when $n \to +\infty$. Let ω_0 be a point where the minimum ψ_0 of ψ is reached and let I_n be the interval $I_n = [\omega_0 - 1/n, \omega_0 + 1/n]$; if we consider the functions u_n such that

$$\tilde{u}_n(\omega) = \begin{cases} (2\pi)^{\frac{1}{2}}\epsilon\left(\int_{I_n} \psi(\omega)\,\mathrm{d}\omega\right)^{-\frac{1}{2}}, & |\omega-\omega_0| \leqslant 1/n, \\ 0, & |\omega-\omega_0| > 1/n, \end{cases} \tag{A 4}$$

then it is easy to verify that

$$\frac{1}{2\pi}\int_{-\infty}^{+\infty}\psi(\omega)\,|\tilde{u}_n(\omega)|^2\,\mathrm{d}\omega = \epsilon^2,\quad \frac{1}{2\pi}\int_{-\infty}^{+\infty}|\tilde{u}_n(\omega)|^2\,\mathrm{d}\omega = \frac{m(I_n)\,\epsilon^2}{\int_{I_n}\psi(\omega)\,\mathrm{d}\omega}, \tag{A 5}$$

where $m(I_n) = 2/n$ is the measure of I_n. Then from the mean value theorem it follows that $\|u_n\| \to \epsilon\psi_0^{-\frac{1}{2}}$ when $n \to +\infty$, and the lemma is proved.

To prove the validity of equation (2.13), remark that the constraint (2.12) is of the type (A 1) with

$$\psi(\omega) = \pi/\cosh \pi\omega + (\epsilon/E)^2\,(\omega^2 + \tfrac{1}{4}). \tag{A 6}$$

Remark also that ψ is an even function of ω and that its minimum for $\omega > 0$ is reached at the point ω_0, which is the unique positive solution of the equation $\psi'(\omega_0) = 0$, i.e.

$$\pi^2 \sinh \pi\omega_0/\cosh^2 \pi\omega_0 = 2(\epsilon/E)^2\,\omega_0. \tag{A 7}$$

It is easy to derive from equation (A 7) that $\omega_0 \to +\infty$ when $\epsilon/E \to 0$; as a consequence, for small values of ϵ/E, equation (A 7) is approximately equivalent to the equation $\pi/\cosh \pi\omega_0 = (2/\pi)\,(\epsilon/E)^2\,\omega_0$, so that $\psi(\omega_0) \sim (\epsilon/E)^2\,\omega_0^2$. It follows that

$$M_0(\epsilon, E) = \sup_{u \in S_0} \|u\| \sim E/\omega_0. \tag{A 8}$$

Finally, from equation (A 7), an elementary argument shows that

$$\omega_0 \sim (2/\pi)\,|\ln(\epsilon/E)|,$$

and the validity of equation (2.13) is proved.

As regards the estimate (2.16), remark that the constraint (2.15) is of the type (A 1) with

$$\psi(\omega) = \pi/\cosh \pi\omega + (\epsilon/E)^2 \cosh 2\alpha\omega. \tag{A 9}$$

Again ψ is an even function of ω and its minimum for $\omega > 0$ is reached at the point ω_0 such that

$$\pi^2 \sinh \pi\omega_0/\cosh^2 \pi\omega_0 = 2\alpha\,(\epsilon/E)^2 \sinh 2\alpha\omega_0. \tag{A 10}$$

We have again that $\omega_0 \to +\infty$ when $\epsilon/E \to 0$ and therefore for small values of ϵ/E we get

$$\frac{\pi}{\cosh \pi\omega_0} \sim \frac{2\alpha}{\pi}\left(\frac{\epsilon}{E}\right)^2 \sinh 2\alpha\omega_0, \tag{A 11}$$

$$\psi_0 = \psi(\omega_0) \sim \frac{1}{2}\left(1 + \frac{2\alpha}{\pi}\right)\left(\frac{\epsilon}{E}\right)^2 \mathrm{e}^{2\alpha\omega_0}, \tag{A 12}$$

$$\omega_0 \sim \frac{1}{\pi + 2\alpha}\ln\left[\frac{\pi^2}{2\alpha}\left(\frac{E}{\epsilon}\right)^2\right]. \tag{A 13}$$

Inserting equations (A 12) and (A 13) in equation (A 2) we get the estimate (2.16).

Appendix B

The results on the eigenvalues of the operator K^*K, quoted in § 3, can easily be derived from standard results on the eigenvalues of a self-adjoint Toeplitz operator

$$(Tf)(x) = \int_{-A}^{+A} T(x-y)f(y)\,\mathrm{d}y, \quad |x| \leqslant A, \tag{B 1}$$

where $T^*(x) = T(-x)$. Indeed, the eigenvalue equation (3.8) can be written in the form

$$\int_a^b K\left(\frac{t}{s}\right) k_k(s)\frac{\mathrm{d}s}{s} = \alpha_k^2 u_k(t), \quad a \leqslant t \leqslant b, \tag{B 2}$$

where

$$K(t) = 1/(1+t). \tag{B 3}$$

With the introduction of the new variables

$$x = \ln[t/(ab)^{\frac{1}{2}}], \quad y = \ln[s/(ab)^{\frac{1}{2}}] \tag{B 4}$$

and the new functions

$$T(x) = \mathrm{e}^{\frac{1}{2}x} K(\mathrm{e}^x), \quad \phi_k(x) = \mathrm{e}^{\frac{1}{2}x} u_k((ab)^{\frac{1}{2}}\mathrm{e}^x), \tag{B 5}$$

equation (B 2) becomes

$$\int_{-A}^{+A} T(x-y)\,\phi_k(y)\,\mathrm{d}y = \alpha_k^2\,\phi_k(x), \quad |x| \leqslant \mathrm{A}, \tag{B 6}$$

where

$$A = \tfrac{1}{2}\ln(b/a) = \tfrac{1}{2}\ln\gamma. \tag{B 7}$$

Therefore the eigenvalue problem (B 2) is equivalent to an eigenvalue problem for a Toeplitz operator. Also, the Fourier transform of the kernel $T(x)$ (equation (B 5)) coincides with the Mellin transform of the kernel $K(t)$; indeed, using equation (B 5), we obtain

$$\hat{T}(\omega) = \int_{-\infty}^{+\infty} T(x)\,\mathrm{e}^{\mathrm{i}\omega x}\,\mathrm{d}x = \int_0^{+\infty} K(t)\,t^{-\frac{1}{2}+\mathrm{i}\omega}\,\mathrm{d}t = \tilde{K}(\omega). \tag{B 8}$$

Here the relation between the variables t, x is $x = \ln t$. In particular, for the kernel (B 3), by means of an elementary computation we get

$$\hat{T}(\omega) = \tilde{K}(\omega) = \pi/\cosh\pi\omega. \tag{B 9}$$

We can now apply the following result on Toeplitz operators:

Theorem B 1. (Gori & Palma 1975). *The eigenvalues of the operator* (B 1) *are non-degenerate if the kernel*

$$H(x) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} \omega\hat{T}'(\omega)\,\mathrm{e}^{-\mathrm{i}\omega x}\,\mathrm{d}\omega \tag{B 10}$$

is definite.

From equation (B 9) it follows that this condition is satisfied since

$$\omega\hat{T}'(\omega) = -\pi^2\omega\sinh\pi\omega/\cosh^2\pi\omega \leqslant 0 \tag{B 11}$$

and therefore property (i) of § 3 is proved.

The following extension of a theorem of Szegö can also be used:

THEOREM B 2 (Miller 1974; Kac *et al.* 1953). *If* $N_A(\delta_0, \delta_1)$ *is the number of eigenvalues of* (B 1) *falling within* (δ_0, δ_1), *if* (δ_0, δ_1) *does not contain* 0 *and if* $\mu(\delta_0, \delta_1)$ *is the measure of the set where* $\delta_0 < \hat{T}(\omega) < \delta_1$, *then*

$$\lim_{A\to+\infty} \frac{1}{2A} N_A(\delta_0, \delta_1) = \frac{1}{2\pi}\mu(\delta_0, \delta_1) \tag{B 12}$$

provided the sets where $\hat{T}(\omega) = \delta_0$ *or* $\hat{T}(\omega) = \delta_1$ *are of measure* 0.

All the conditions of the theorem are satisfied by the kernel (B 9): in particular, since $\hat{T}(\omega) > 0$, we can always assume $\delta_0 > 0$. Finally, if we remark that

$$\hat{T}(\omega) = \pi/\cosh \pi\omega = \hat{K}(\omega).$$

is an even function of ω, and that it is a decreasing function for $\omega > 0$, we reach the conclusion that the ω-set, where the condition $\delta_0 < \hat{T}(\omega) < \delta_1$ is satisfied, is the union of two intervals. Using this fact and equation (B 7), we can derive equation (3.12) from equation (B 12).

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