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On the recovery and resolution of exponential relaxation rates from experimental data II. The optimum choice of experimental sampling points for Laplace transform inversion

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In a previous paper (Bertero, Boccacci & Pike, *Proc. R. Soc. Lond.* A **383**, 15 (1982)) we investigated the benefits of *a priori* knowledge of finite support of the solution in the inversion of the Laplace transform and determined the number of exponential components that could be recovered from given data in the presence of quantified amounts of noise. The support of the data was assumed to be unrestricted. In the present paper these calculations are extended to cover the case of a necessarily finite number of experimental data points. We consider two cases: uniform distribution and geometric distribution of data points. In both cases the condition number of singular value inversions is minimized with respect to the placing of the data points for fixed numbers of data samples. The results allow optimum placing of the experimental data points as a function of the ratio, γ, of given upper and lower bounds of the support of the solution. The number of exponentials recoverable is less than in the unrestricted case but for $\gamma \leqslant 8$ we find that with 32 linearly spaced data points, optimally placed, this reduction is rather small. Geometrical sampling of experimental data reduces further the number of sample points required for inversion. For $\gamma \leqslant 8$ we show that with 5 geometrically spaced data points, points, optimally placed, the ill-conditioning in the restoration of 2 to 4 exponential components is smaller than with 32 linearly spaced data samples. This has important implications for the design of a number of scientific experiments and instruments.

1. Introduction

In Bertero *et al.* (1982) the problem of numerical inversion of the Laplace transform

$$g(p) = \int_0^{+\infty} \mathrm{e}^{-pt} f(t)\,\mathrm{d}t, \quad 0 \leqslant p < \infty, \tag{1.1}$$

was considered. The 'solution' $f(t)$ was assumed to be of known, strictly positive support while the 'data' $g(p)$, albeit corrupted by additive Gaussian noise, were assumed to be known everywhere.

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A first step in the investigation of this problem by means of information theoretical methods had been taken by McWhirter & Pike in 1978. Their analysis applies to the case where no *a priori* knowledge on the support of the unknown function $f(t)$ is available. Their results can be summarized as follows: it is impossible to resolve $f(t)$ between any point t and a second point $t' > t$ unless

$$t' \geqslant \delta_0 t, \quad \delta_0 = \exp(\pi/\omega_0), \tag{1.2}$$

where ω_0 is the greatest value, for a given signal-to-noise ratio, of a generalized frequency ω at which components may be extracted from $g(p)$.

This result provides a basis for a practical method of solution of (1·1) (Ostrowsky *et al.* 1981); indeed, if the unknown function $f(t)$ is uniformly sampled in the variable $\ln t$ and if the sampling points are taken at a rate in agreement with (1.2), then it is to be expected that wildly oscillating solutions are excluded and that a regularization through discretization of the ill-posed problem (1.1) is obtained.

An interesting result, however, derived by Ostrowsky *et al.* (1981) by means of numerical simulations is that resolution beyond the limit (1.2) is possible when $f(t)$ has a bounded (strictly positive) support. In such a case (1.1) takes the form

$$g(p) = \int_a^b e^{-pt} f(t)\,dt, \quad 0 \leqslant p < \infty, \tag{1.3}$$

where a (> 0) and b ($> a$) are, respectively, the lower and the upper limits of the support of $f(t)$.

The improvement in resolution discovered by Ostrowsky *et al.* (1981) was theoretically investigated by Bertero *et al.* (1982) by means of singular function expansions, which provide an extremely powerful approach to the solution of first kind Fredholm integral equations (Miller 1974). Singular values and singular functions of the 'finite Laplace transform' (1.3) were investigated and the distribution of the zeros of singular functions was used to define a 'resolution ratio' δ_r, which is a function of the signal to noise ratio E/ϵ and of the parameter, characterizing the support of $f(t)$,

$$\gamma = b/a \ (> 1). \tag{1.4}$$

In particular it was proved that $\delta_s < \delta_0$, δ_0 being the resolution ratio of (1.2), that $\delta_s \to 1$ when $\gamma \to 1$ and that $\delta_s \to \delta_0$ when $\gamma \to \infty$. We wish also to emphasize that the 'asymptotic limit' δ_0 is only approached when $\ln \gamma$ is very large, therefore even a rough estimation of a and b can allow a significant improvement beyond the resolution limit δ_0.

However, the main result derived in Bertero *et al.* (1982) is that they determined how many generalized Fourier components of $f(t)$ could be extracted from a noisy data function $g(p)$, for a given *a priori* knowledge of the signal to noise ratio and of the support of $f(t)$. Let K_s be this maximum number of 'Fourier components'; then K_s is a generalized 'number of degrees of freedom' (Shannon number) or also a measure of the 'amount of information' contained in $g(p)$. The relation between the number of degrees of freedom K_s and the resolution ratio δ_s is given simply by:

$$\delta_s^{K_s} = \gamma. \tag{1.5}$$

In these previous investigations it was assumed that $g(p)$ is known for all values of p in $(0, +\infty)$. But, as is obvious, in practical problems $g(p)$ is known only on a finite set of points, so that (1.3) should be replaced by

$$g(p_n) = \int_a^b e^{-p_n t} f(t)\,dt; \quad n = 1, \ldots, N. \tag{1·6}$$

Then in order to apply to this case the theoretical results derived in Bertero *et al.* (1982), the following problem has to be solved: if K_s is the number of degrees of freedom corresponding to given values of the signal to noise ratio and of the parameter γ, find a set of points $\{p_n\}_{n=1}^N$ which allow an accurate determination of the first K_s components of $g(p)$. The exact solution of this problem requires accurate computation of the singular functions and singular values of the 'finite Laplace transform' (1.6) over a range of values of γ, with the number and spacing of data points as parameters. While such calculations are not out of the question, we investigate for the present a more 'practical' approach to the problem of the optimum choice of the data sampling points $\{p_n\}_{n=1}^N$.

We assume a 'model', dependent on a small number of parameters (of the same order as the number of degrees of freedom), for the unknown function $f(t)$ and we restrict the distribution of the sampling points p_n of the experimental data to the uniform distribution

$$p_n = p_1 + d(n-1); \quad n = 1, 2, \ldots, N \tag{1.7}$$

or to the geometric distribution ('exponential sampling' of data: Pike *et al.* 1982).

$$p_n = p_1 \Delta^{n-1} \quad n = 1, 2, \ldots, N. \tag{1.8}$$

For a given 'model' and a given distribution of the sampling points, the parameters of the model can be determined by solving a least squares problem. This is usually ill conditioned and its condition number will depend on the distribution of the data sampling points. Then an 'optimum' distribution is obtained by minimizing the condition number. In this way, for a given 'model' for $f(t)$ and a given number N of data sampling points, we can find optimum values of $\{p_1, d\}$ in the case of uniform distribution and optimum values of $\{p_1, \Delta\}$ in the case of geometric distribution.

As concerns the choice of the model for $f(t)$, in the next section we justify the use of the 'exponential sampling' model of Ostrowsky *et al.* (1981).

2. The exponential sampling model

In Bertero *et al.* (1982) – hereafter referred to as I – it was shown that the singular values of the finite Laplace transform (1.3), which defines a compact, injective operator on $L^2(a, b)$ into $L^2(0, \infty)$, depends on the support $[a, b]$ only through the parameter γ, (1.4). Therefore it is convenient, by means of a change of variables, to write (1.6) in the following form

$$g(p_n) = \int_1^\gamma e^{-p_n t} f(t)\,dt; \; n = 1, \ldots, N. \tag{2.1}$$

Since in this paper we deal with the problem of defining and computing an 'optimum set' of sampling points for the experimental data $g(p)$, we note, first, that when an 'optimum set' $\{p_1, \ldots, p_N\}$ is found for the problem (2.1), then the 'optimum set' for the problem (1.6) is given by $\{p_1/a, \ldots, p_N/a\}$. Thus the dependence of the best sampling points on the lower bound of the support of $f(t)$ is extremely simple.

From the results derived in I it follows that, given γ and given the signal-to-noise ratio E/ϵ, it is possible to determine the number of components K_s of the unknown function $f(t)$ that can be recovered from the noisy data $g(p)$. The 'number of degrees of freedom' K_s is defined as the number of singular values that are greater than ϵ/E. In other words, we cannot completely recover the function $f(t)$ but only the projection $\tilde{f}(t)$ of $f(t)$ on a subspace with dimension K_s. It must also be noted that the dimension of this subspace is usually rather small. For instance, in the case $\gamma = 5$, only 3 singular values are greater than 10^{-2} and only 5 singular values are greater than 10^{-3}. A signal-to-noise ratio of the order of 10^3 is the best value achievable in many experimental situations (Pike *et al.* 1982).

From the results of I and of Ostrowsky *et al.* (1981), it follows that the projection $\tilde{f}(t)$ of $f(t)$ can be approximated by a known combination of K_s ln (sinc) functions, centred at points t_k, forming a geometric progression with ratio δ_s, as given by (1.5). The coefficients of the linear combination, namely the amplitudes of the ln (sinc) functions, can be computed by solving a least squares problem. In Ostrowsky *et al.* (1981) it is also shown that no significant error in the determination of the coefficients results if the ln (sinc) functions are replaced by delta functions (point masses) centred at the same points.

Thus, the following computational scheme for $\tilde{f}(t)$ is suggested, analogous to the Fourier sampling expansion: (i) given γ and E/ϵ, determine K_s and δ_s; (ii) consider partition of the interval $[1, \gamma]$ into K_s adjacent sub-intervals $J_k = [\delta_s^{k-1}, \delta_s^k]$ $(k = 1, \ldots, K_s)$; (iii) locate a delta function in each J_k and determine the coefficients by solving a least squares problem; (iv) use the coefficients of the delta functions as coefficients of a linear combination of ln (sinc) functions (interpolation scheme).

We can call any subinterval J_k a 'resolution element'. Then the location of each delta function inside a given 'resolution element' J_k is not very important; it is only important that the positions t_k of the delta functions form a geometric progression with ratio δ_s. Indeed, as follows from the theory of the resolving power of Laplace inversion developed in I and in McWhirter & Pike (1978), it is impossible to resolve two 'point masses' that lie inside the same 'resolution element'. We also notice that the problem of obtaining the maximum number of 'point masses' consistent with the data has a very simple solution. This number is just K_s and, as follows from our computations of the singular values of the finite Laplace transform, it is not surprising, contrary to what was believed by Ruhe (1980), for example, that this number is always quite small.

From the previous remarks it follows that the 'exponential sampling model' of Ostrowsky *et al.* (1981) provides a practical and efficient way for determining the projection $\tilde{f}(t)$ of the unknown solution $f(t)$ and therefore it can also be used in the

analysis of the problem (2.1) with discrete data. For the same reasons we believe that our procedure for the choice of the data points p_n is not very sensitive to the model. Thus, once the 'optimum set' of data points has been determined in the case of the 'exponential sampling model', this set can be used in any method of inversion, for example in the histogram model of Gulari *et al.* (1979) in the cubic B-spline model of McWhirter (1980, 1981) or in more constrained ones, for example, the nonlinear fitting method described by Provencher (1979).

We give now a description of our procedure: let us consider a partition of the interval $[1, \gamma]$ into K adjacent subintervals $J_k = [\delta^{k-1}, \delta^k]$ $(k = 1, 2, \ldots, K)$ where

$$\delta^K = \gamma; \tag{2.2}$$

in each interval points t_k are taken such that

$$t_k = \delta^{k-1} t_1; \quad k = 1, \ldots, K; \quad 1 < t_1 < \delta. \tag{2.3}$$

For instance, in the case $t_1 = \frac{1}{2}(1+\delta)$, the points t_k are the middle points of the intervals J_k and they are given by

$$t_k = \tfrac{1}{2}(1+\delta)\delta^{k-1}; \quad k = 1, \ldots, K. \tag{2.4}$$

For a given set of points t_k one considers the 'function'

$$\check{f}(t) = \sum_{k=1}^{K} c_k \, \delta(t - t_k) \tag{2.5}$$

(as shown in Ostrowsky *et al.* (1981) the coefficients c_k are approximately proportional to the values of the restored solution $\tilde{f}(t)$ at the points t_k) and the corresponding Laplace transform

$$\check{g}(p) = \sum_{k=1}^{K} c_k \, \mathrm{e}^{-pt_k}; \tag{2.6}$$

the unknown parameters c_k are determined by solving the least squares problem

$$\sum_{n=1}^{N} w_n |g(p_n) - \check{g}(p_n)|^2 = \text{minimum}, \tag{2.7}$$

where $\{g(p_n)\}$ is the set of the data and the weights w_n are introduced in order to take into account a non-uniform distribution of the sampling points. If we use, for instance, a trapezoidal rule, then

$$\left.\begin{aligned} &w_n = \tfrac{1}{2}(p_{n+1} - p_n); \quad n = 1; \qquad w_n = \tfrac{1}{2}(p_n - p_{n-1}); \quad n = N, \\ &w_n = \tfrac{1}{2}(p_{n+1} - p_{n-1}); \quad n = 2, \ldots, N-1. \end{aligned}\right\} \tag{2.8a}$$

Other choices that we shall use are suitable for integrated sampling, namely,

$$w_n = \text{constant, for the case of uniform sampling, and,} \tag{2.8b}$$

$$w_n = \text{constant} \times p_n \text{ for the case of geometric sampling.} \tag{2.8c}$$

The success of this procedure is strictly related to the ill-conditioning of problem (2.7), whose solution is equivalent to the solution of the Euler equation

$$[A_w^{\mathrm{T}} A]\,\boldsymbol{c} = [A_w^{\mathrm{T}}]\,\boldsymbol{g}. \tag{2.9}$$

Here $\boldsymbol{c}$ is the K-dimensional vector of the coefficients c_k, $\boldsymbol{g}$ is the N-dimensional vector of the data $g(p_n)$, while the matrix elements of $[A]$ and $[A_w^{\mathrm{T}}]$ are given by

$$\left.\begin{aligned} [A]_{nk} &= \mathrm{e}^{-p_n t_k}, \\ [A_w^{\mathrm{T}}]_{kn} &= w_n\,\mathrm{e}^{-p_n t_k} \end{aligned}\right\} k = 1, \ldots, K;\ n = 1, \ldots, N. \tag{2.10}$$

Now, if we denote by $\beta_1, \ldots, \beta_K$ the square roots of the eigenvalues of the matrix $[A_w^{\mathrm{T}} A]$ and by $\boldsymbol{u}^{(1)}, \ldots, \boldsymbol{u}^{(K)}$ the corresponding eigenvectors, then the solution of (2.9) can be expressed as follows:

$$\boldsymbol{c} = \sum_{k=1}^{K} c_k \boldsymbol{u}^{(k)} = \sum_{k=1}^{K} \frac{1}{\beta_k} (\boldsymbol{g}, \boldsymbol{v}^{(k)})\, \boldsymbol{u}^{(k)}, \tag{2.11}$$

where $\boldsymbol{v}^{(k)} = \beta_k^{-1}[A]\,\boldsymbol{u}^{(k)}$ and $(\boldsymbol{g}, \boldsymbol{v}^{(k)})$ is the weighted scalar product

$$(\boldsymbol{g}, \boldsymbol{v}^{(k)}) = \sum_{n=1}^{N} w_n g(p_n)\, (\boldsymbol{v}^{(k)})_n. \tag{2.12}$$

Therefore, if we introduce the condition number

$$\beta^{(K)} = \beta_1/\beta_K \tag{2.13}$$

we obtain the well known inequality

$$\|\delta \boldsymbol{c}\|/\|\boldsymbol{c}\| \leqslant \beta^{(K)} \|\delta \boldsymbol{g}^{(K)}\|/\|\boldsymbol{g}^{(K)}\|, \tag{2.14}$$

which describes propagation of errors in the solution of problem (2.7). Here $\|\boldsymbol{c}\|$ is the usual Euclidean norm in K-dimensional space while $\|g\|$ is the norm induced by the weighted scalar product (2.12); $\boldsymbol{g}^{(K)}$ denotes the projection of $\boldsymbol{g}$ on the subspace spanned by the singular vectors $\boldsymbol{v}^{(1)}, \ldots, \boldsymbol{v}^{(K)}$.

The condition number $\beta^{(K)}$ can easily be evaluated by computing the square roots of the greatest and smallest eigenvalues of the $K \times K$ matrix $[A_w^{\mathrm{T}} A]$; this is not a tremendous task since, as remarked before, the interesting values of K usually are rather small. For a given set $\{p_n\}_{n=1}^{N}$ of data points and a given set $\{t_k\}_{k=1}^{K}$ of solution points, the matrix elements of $[A_w^{\mathrm{T}} A]$ are given by

$$[A_w^{\mathrm{T}} A]_{ik} = \sum_{n=1}^{N} w_n\,\mathrm{e}^{-p_n(t_i+t_k)}. \tag{2.15}$$

In the case of uniform sampling, taking $w_n = d$ $(n = 1, \ldots, N)$, (2.8*b*), the right side of (2.15) takes the simple form

$$[A_w^{\mathrm{T}} A]_{ik} = d\,\mathrm{e}^{-p_1(t_i+t_k)}[1 - \mathrm{e}^{-Nd(t_i+t_k)}]/[1 - \mathrm{e}^{-d(t_i+t_k)}]. \tag{2.16}$$

For given values of γ, K and N the condition number $\beta^{(K)}$ depends on the parameters $\{p_1, d\}$, and we define as an 'optimum choice' of these parameters the values that minimize the condition number $\beta^{(K)}$.

Analogously if we consider the geometric distribution (1.8) of data points and if we choose the weights w_n according to (2.8a), for example,

$$\left.\begin{aligned} &w_n = \tfrac{1}{2}p_1\Delta^{n-1}(\Delta-1); \quad n=1; \quad w_n = \tfrac{1}{2}p_1\Delta^{n-2}(\Delta-1); \quad n=N, \\ &w_n = \tfrac{1}{2}p_1\Delta^{n-2}(\Delta^2-1); \quad n=2,\ldots,N-1, \end{aligned}\right\} \tag{2.17}$$

then, for given values of γ, K and N the condition number $\beta^{(K)}$ depends on the parameters $\{p_1, \Delta\}$ and also in this case we define as an 'optimum choice' of these parameters the values that minimize the condition number $\beta^{(K)}$.

3. Optimum choice of experimental sampling points: numerical results

Since the interesting values of K are not very large (for $\gamma \leqslant 5$ and $E/\epsilon \leqslant 10^3$, K_s does not exceed 5) the condition number of the $K \times K$ matrix (2.15) can be easily computed by means of a desk computer, and the procedure described in § 2 can be put into practice. Notice that bounds on the condition number can sometimes be obtained by the results of Ruhe (1980).

In the case of uniform sampling it is easy to verify by numerical computations that, for fixed d, the condition number of the matrix (2.16) is an increasing function of p_1 so that the minimum is reached at $p_1 = 0$. Since in many practical experiments the Laplace transform cannot be measured at the point $p_1 = 0$, the previous result indicates that it is necessary to take p_1 as small as possible. In practice that means $p_1 = d$. We have verified that, for $d < 0.1$, the difference between the condition number of matrix (2.16) with $p_1 = 0$ and the condition number of the matrix (2.16) with $p_1 = d$ is rather small (less than a few per cent).

In the case $p_1 = d$, the matrix (2.16) takes the form

$$[A_w^{\mathrm{T}}A]_{ik} = d\frac{1-\exp[-Nd(t_i+t_k)]}{\exp[d(t_i+t_k)]-1}; \quad i,k=1,\ldots,K, \tag{3.1}$$

the points t_k being defined by (2.4) and (2.2); therefore, for fixed γ, K, N, we have to minimize the condition number with respect to only one parameter, namely the distance d between data sampling points. We denote by $\beta^{(K)}(d)$ the square root of the condition number of matrix (3.1); see also (2.13).

It is easy to recognize, by considering the approximate values of the matrix elements (3.1) in the limits $d \to 0$ and $d \to +\infty$, that $\beta^{(K)}(d) \to +\infty$ both when $d \to 0$ and when $d \to +\infty$. As a consequence, the function $\beta^{(K)}(d)$ has certainly a minimum. It follows from numerical computations that in fact only one minimum exists; we denote by $\hat{\beta}^{(K)} = \beta^{(K)}(d_0)$ the minimum value of $\beta^{(K)}(d)$ at the minimum point $d = d_0$. We also notice that the matrix (3.1) has a limit when $N \to \infty$

$$[A_w^{\mathrm{T}}A]_{ik} = \frac{d}{\exp[d(t_i+t_k)]-1}, \tag{3.2}$$

and that the condition number of this matrix is also a function of d. In this case

$\beta^{(K)}(d)$ does not tend to infinity when $d \to 0$ since for $d = 0$ the matrix (3.2) takes the form

$$[A_w^{\mathrm{T}} A]_{ik} = 1/(t_i + t_k), \tag{3·3}$$

but it is still true that $\beta^{(K)}(d) \to +\infty$ when $d \to +\infty$.

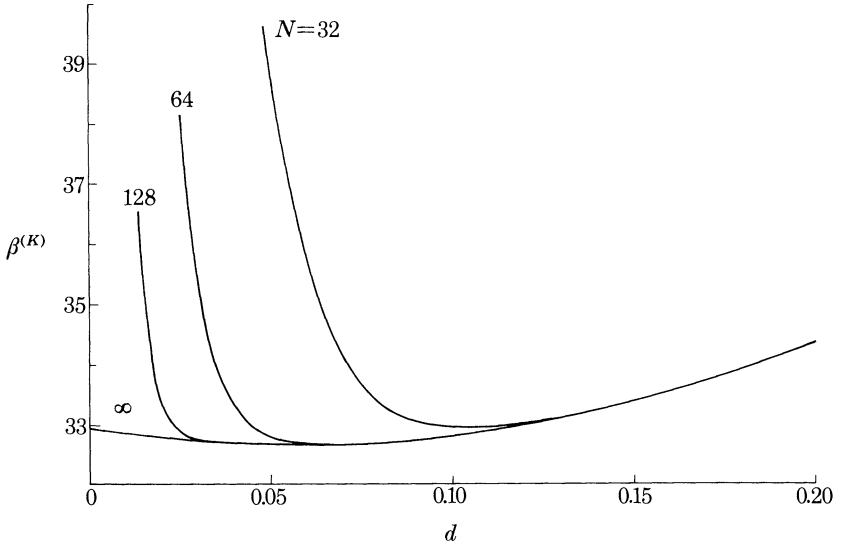

FIGURE 1. The behaviour of the square root of the condition number of matrix (3.1) as a function of the distance d between data points, for various values of the number N of data points. Here $\gamma = 5$ and $K = 3$.

Computations have been performed for $K = 2$–5, $\gamma = 1.5$–8 and for $N = 32$, 64, 128, 256, ∞. An interesting result is that, in the range of values of γ and K we have considered, the values of $\hat{\beta}^{(K)}$ are practically independent of N and therefore $N = 32$ is an adequate value for the number of data points. The result is illustrated by an example in figure 1 where we represent the behaviour of $\beta^{(K)}(d)$ as a function of d in the case $\gamma = 5$, $K = 3$ and for various values of N. Note that the condition number of the matrix (3.2) does not take its minimum value at $d = 0$, even if the behaviour of $\beta^{(K)}(d)$ in the neighbourhood of $d = 0$ is very flat. Indeed we have $\beta^{(K)}(0) = 32.964$ while the value $\beta^{(K)}(d)$ at the minimum point $d = d_0 = 0.05605$ is $\hat{\beta}^{(K)} = \beta^{(K)}(d_0) = 32.678$.

As clearly appears from figure 1, the graph of the function $\beta^{(K)}(d)$ corresponding to $N = +\infty$ is the envelope of the graphs of the functions $\beta^{(K)}(d)$ corresponding to finite values of N. Therefore a typical behaviour of these functions is that they grow very rapidly to the left of the minimum while they grow rather slowly to the right. Furthermore, by increasing N, the value d_0 decreases (the limiting value is the value of d_0 for the matrix (3·2) and the minimum becomes more and more flat (an indication of oversampling of data)) while, as we already remarked, the minimum value of $\beta^{(K)}(d)$ does not change significantly. Even if the value of d_0 decreases for increasing

N, the value $L_0 = Nd_0$, which represents the cut-off in the measurement of the Laplace transform, increases and, when N is sufficiently large, it is approximately proportional to N. This behaviour is shown in table 1.

TABLE 1. BEHAVIOUR OF d_0, L_0 AND $\hat{\beta}^{(k)}$ AS A FUNCTION OF K AND N IN THE CASE $\gamma = 5$

(The values of $\alpha^{(K)}$ for $\gamma = 5$ are: $\alpha^{(2)} = 4.52$, $\alpha^{(3)} = 22.9$, $\alpha^{(4)} = 118$, $\alpha^{(5)} = 610$.)

	$K = 2$			$K = 3$			$K = 4$			$K = 5$		
N	d_0	L_0	$\hat{\beta}^{(2)}$	d_0	L_0	$\hat{\beta}^{(3)}$	d_0	L_0	$\hat{\beta}^{(4)}$	d_0	L_0	$\hat{\beta}^{(5)}$
32	0.12	3.84	5.35	0.10	3.20	33.0	0.12	3.84	215	0.14	4.48	1453
64	0.12	7.68	5.35	0.07	4.48	32.7	0.07	4.48	209	0.08	5.12	1367
128	0.12	15.4	5.35	0.06	7.68	32.7	0.04	5.12	207	0.05	6.40	1345
256	0.12	30.7	5.35	0.06	15.4	32.7	0.04	10.2	207	0.03	7.68	1341
∞	0.12	∞	5.35	0.06	∞	32.7	0.04	∞	207	0.03	∞	1341

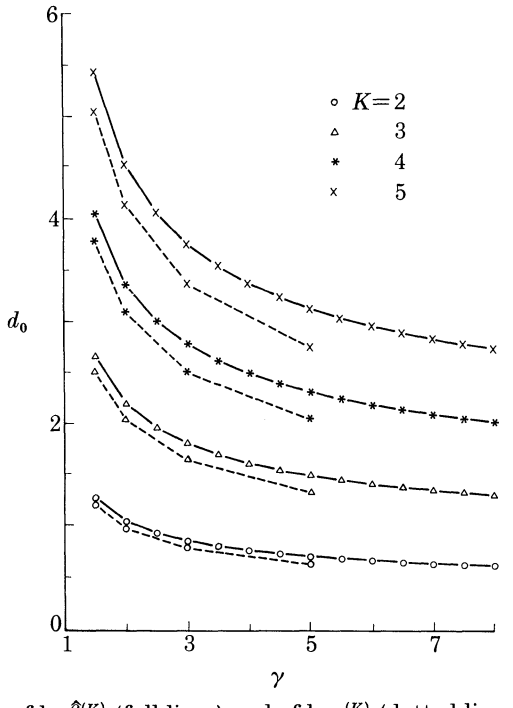

FIGURE 2. The behaviour of $\lg \hat{\beta}^{(K)}$ (full lines) and of $\lg \alpha^{(K)}$ (dotted lines) as a function of γ, for various values of K and in the case of 32 uniformly spaced data points.

In figure 2 we give the behaviour of $\lg \hat{\beta}^{(K)}$ as a function of γ (full lines) in the case of 32 data points. The behaviour of $\lg \alpha^{(K)}$ is given in the same figure (dotted lines), $\alpha^{(K)}$ being defined by the ratio of the first to the Kth singular values of the problem with continuous data. This behaviour is derived from the singular values computed in Bertero *et al.* (1982). It may be seen that the difference between $\hat{\beta}^{(K)}$ and $\alpha^{(K)}$ is not very large for small K; for $K \geqslant 4$, we have approximately $\hat{\beta}^{(K)} \sim 2\alpha^{(K)}$. We see that

the ratio $\hat{\beta}^{(K)}/\alpha^{(K)}$ grows for increasing values of K and the previous investigation of the limit $N \to \infty$ shows that this fact has to be attributed mainly to the 'exponential sampling' model (or to any similar model) rather than to an inadequate number of data points. Indeed, as we have already recalled, we have found no significant change in $\hat{\beta}^{(K)}$ when $N \geqslant 32$.

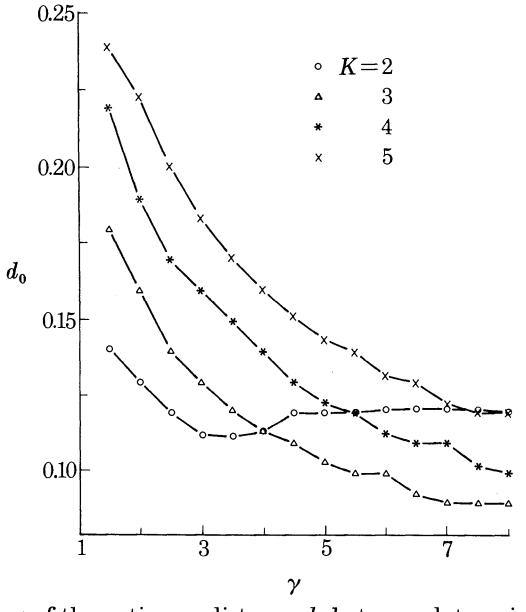

FIGURE 3. The behaviour of the optimum distance d_0 between data points as a function of γ, for various values of K and in the case of 32 uniformly spaced data points.

In figure 3 we give the behaviour of d_0, the 'optimum' distance between data points, as a function of γ and for $K = 2$–5, always in the case of 32 data points. In fact, the minimum of $\beta^{(K)}(d)$ is rather flat and hence it is useful to have not only the value of d_0 but also a range of values of d such that $\beta^{(K)}(d)$ does not differ significantly from $\hat{\beta}^{(K)}$. We illustrate this by an example in figure 4 where we give the behaviour of $\lg \beta^{(K)}(d)$ (in the case $K = 3$) as a function of d for various values of γ. As we see the range of admissable values of d decreases for increasing γ.

In the case of geometric sampling, (1.8), the matrix elements $[A_w^{\mathrm{T}} A]_{ik}$ can be computed only by means of (2.15) and the minimization of the condition number has to be performed with respect to two parameters, the position p_1 of the first data point and the dilation factor Δ. Computations are therefore more time consuming than in the case of uniform sampling. However, from theoretical considerations founded on the sampling theorem (Pike *et al.* 1982) it is expected that the number of data points required in the case of geometric sampling is much smaller than the number of data points required in the case of uniform sampling. For these reasons we have performed numerical computations mainly in the case of 5 data points, which is the smallest number of data points required for the restoration of 5 exponential components.

We will denote as $\beta^{(K)}(p_1)$ the square root of the condition number of the matrix $A_w^{\mathrm{T}} A$ for fixed Δ and as $\hat{\beta}^{(K)} = \beta^{(K)}(p_{1,0})$ the minimum value of $\beta^{(K)}(p_1)$ at the minimum point $p_{1,0}$. For all the values of Δ we have considered, the function $\beta^{(K)}(p_1)$ has two local minima and the smallest value is that corresponding to the local minimum to

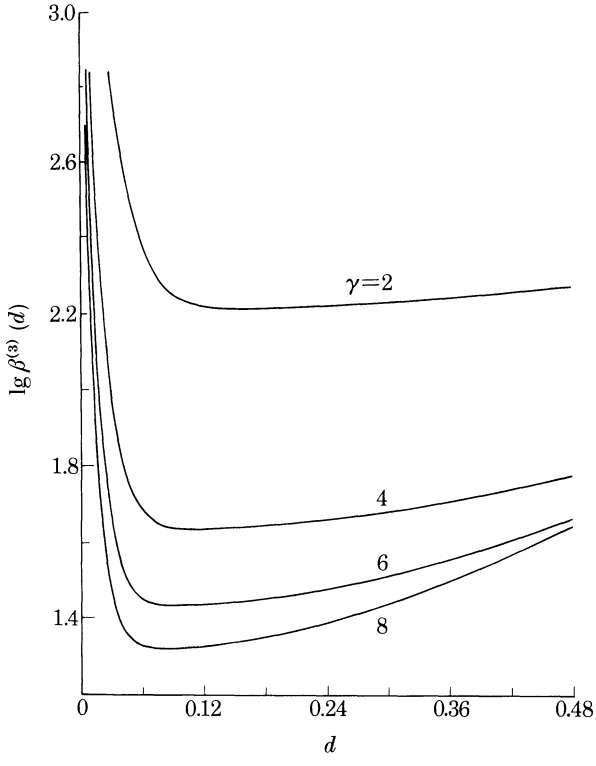

FIGURE 4. The behaviour of $\lg \beta^{(K)}(d)$, with $K = 3$, as a function of d for various values of γ and in the case of 32 uniformly spaced data points.

the right. Such behaviour is illustrated in figure 5 which corresponds to the restoration of three exponentials ($K = 3$) in the case $\gamma = 5$, with five data points of dilation factor $\Delta = 5.5$ (compare with the behaviour of the condition number in the case of uniform sampling, figure 1, for the same values of γ and K). The full line corresponds to the choice of the weights w_n given in (2.17) while the dotted line corresponds to the choice of the weights w_n as in (2.8*c*):

$$w_n = (\ln \Delta) p_1 \Delta^{n-1}: \quad n = 1, \ldots, N, \tag{3.4}$$

which is suggested by considerations founded on the sampling theorem mentioned above. As one can see the behaviour is rather similar in the two cases and the local minimum to the left is approximately the same and at the same value of p_1 in both cases. The main difference is in the neighbourhood of the first local minimum. We may notice that the position of the minimum to the right is obtained approximately

by multiplication of the position of the minimum to the left by Δ. We attribute this behaviour to the dominant significance of the four common points in these two positions in the reduction of the condition number. (We thank a referee for this observation.)

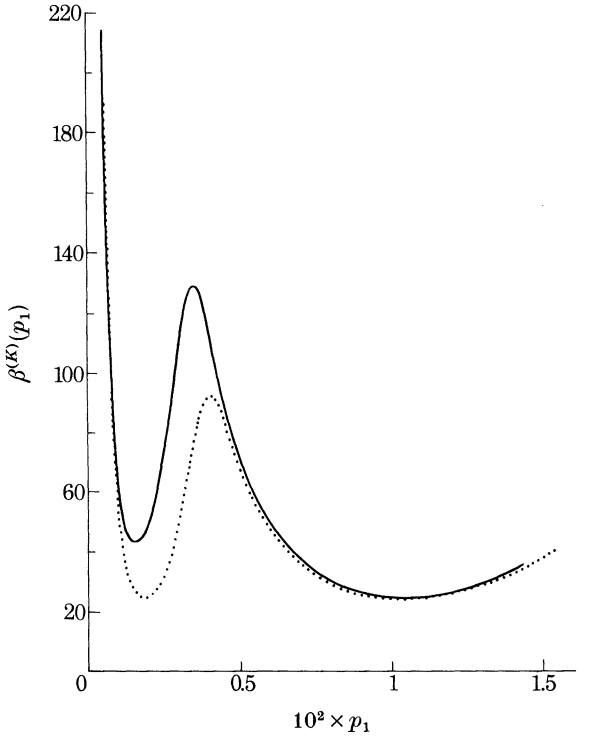

FIGURE 5. The behaviour of the square root of the condition number of matrix (2.15) as a function of p_1, the position of the first data point, in the case of 5 geometrically spaced data points with $\Delta = 5.5$. Here $\gamma = 5$ and $K = 3$. The full line has been computed by using the weights (2.17) and the dotted line by using the weights (3.4).

From a comparison of figure 1 with figure 5 it is immediately apparent that the minimum value of the condition number in the geometric case is smaller than the smallest value of the condition number in the case of uniformly distributed data.

In order to minimize the condition number with respect to the two parameters p_1 and Δ, we have minimized the condition number with respect to p_1 for various fixed values of Δ, starting from $\Delta = 1.4$ and changing Δ by one unit on the second significant digit at each step. We denote as Δ_0 the best value of Δ found in such a way. In table 2 we report the results obtained in the case $K = 3$ with $N = 5$ geometrically spaced points. The matrix elements (2.15) have been computed by using the weights (2.17) and the positions (2.4) of the exponential components. The parameter L_0 is the cut-off in the measured Laplace transform, given by

$$L_0 = p_{1,0}\Delta_0^{N-1} \tag{3.5}$$

(in table 2, $N = 5$). A remarkable feature is that the optimum dilation factor Δ_0 is not very sensitive to γ; furthermore, minima are rather flat in the neighbourhood of Δ_0, $p_{1,0}$. Therefore if we take $\Delta = 5.5$ for all the values of γ in the range $\gamma = 2$–8 and if we minimize with respect to p_1, we obtain minimum values of the condition number which coincide (within 0.1 %), with the values reported in table 2.

TABLE 2. BEHAVIOUR OF Δ_0, $p_{1,0}$, L_0 AND $\hat{\beta}^{(K)}$ AS A FUNCTION OF γ IN THE CASE $K = 3$ AND $N = 5$

(In the last two columns we reflect, for comparison, the values of $\alpha^{(3)}$, the condition number of the continuous problem, which can be computed from the results reported in Bertero *et al.* (1982), and the values of $\beta^{(3)}$ (here denoted as $\beta^{(3)}_{\text{lin}}$) corresponding to 32 linearly spaced data points.)

γ	Δ_0	$p_{1,0}$	L_0	$\beta^{(3)}$	$\alpha^{(3)}$	$\beta^{(3)}_{\text{lin}}$
2	5.4	1.7×10^{-2}	14.4	120	114	165
3	5.4	1.4×10^{-2}	11.8	49.5	46.7	67.3
4	5.5	1.1×10^{-2}	10.5	32.2	—	43.4
5	5.5	1.0×10^{-2}	9.4	24.7	22.9	33.0
6	5.6	8.9×10^{-3}	8.8	20.6	—	27.2
7	5.7	7.9×10^{-3}	8.3	18.0	—	23.5
8	5.8	7.0×10^{-3}	7.9	16.2	—	21.0

The previous remark simplifies computations since we have to minimize with respect to only one parameter, namely p_1. In the case $K = 2$ the average value of Δ is $\Delta = 6$ while in the case $K = 4$ the average value of Δ is $\Delta = 3$. Both in the case $K = 2$ and in the case $K = 4$ we have found, as in the case $K = 3$, minimum condition numbers which are always smaller than the corresponding minimum condition numbers obtained with 32 linearly spaced points.

Finally we notice that, if we look for the restoration of the distribution (2.5) using continuous data $g(p)$, then we have to solve the least squares problem

$$\int_0^\infty \left| g(p) - \sum_{k=1}^{K} c_k \, \mathrm{e}^{-p_k t} \right|^2 \mathrm{d}p = \text{minimum}. \tag{3.6}$$

The associated Euler equation takes the form (2.9) where now the matrix $[A_w^{\mathrm{T}} A]$ is given by (3·3). As we know, in the case $K = 3$ and $\gamma = 5$, the square root of the condition number of this matrix, namely the condition number of problem (3.6), is 32.964 and therefore it is greater than the smallest condition number obtained by using 5 optimally placed geometrically spaced data points (from table 2, $\hat{\beta}^{(3)} = 24.7$).

As a consequence of this result and as indicated by figure 5, there are values of p_1, where $\beta^{(K)}(p_1)$ takes the value 32.964. More precisely, there are two such values of p_1 in the case of the weights (2.17) and four values in the case of the weights (2.4); then, if we compute the singular values of the problem (2.7) in a neighbourhood of these values of p_1 we find surprisingly good approximations of the singular values of the problem (3.6).

As an example we give again the results in the case $K = 3$ and $\gamma = 5$. In this case

the singular values of the problem (3.6) (namely, the square roots of the eigenvalues of the matrix (3.3)) are

$$\beta_1 = 0.827, \quad \beta_2 = 0.163, \quad \beta_3 = 0.0251. \tag{3.7}$$

Then, if we remark that both the functions represented in figure 5 take the value $\beta^{(K)}(0) = 33$ at the point $p_1 = 0.75 \times 10^{-2}$, we compute at this point the singular values of the problem (2.7). In the case of the weights (2.17) we find

$$\beta_1 = 1.017, \quad \beta_2 = 0.201, \quad \beta_3 = 0.0309, \tag{3.8}$$

and, in the case of the weights (3.4).

$$\beta_1 = 0.818, \quad \beta_2 = 0.162, \quad \beta_3 = 0.0247. \tag{3.9}$$

The difference between the singular values (3.7) and the singular values (3.9) does not exceed 1.5 %.

4. Conclusions

From the investigations reported in this paper it follows that an 'optimum choice' of the data points in the Laplace transform inversion can be determined by looking for data point distributions that minimize the condition number of the inversion procedure.

In such a way we have found that the condition numbers of the 'exponential sampling model' proposed in Ostrowsky *et al.* (1981) are always greater than the corresponding condition numbers of the singular value method proposed in Bertero *et al.* (1982), even if the difference is not very large, especially in the case of geometrically spaced data points. It is also important to notice that the singular values of the 'exponential sampling model' provide satisfactory approximations of the singular values of the continuous problem. For instance, in the case $\gamma = 5$, the first three singular values of the latter problem, as computed in I, are $\alpha_0 = 0.875$, $\alpha_1 = 0.193$, $\alpha_2 = 0.0383$ (compare with (3.7)). This result, and similar results for other values of γ, indicates that the 'exponential sampling model' provides an efficient and practical method for Laplace inversion.

Another important result is that, always in the framework of the 'exponential sampling model', the problem with continuous data can be very well approximated by a problem with a very small number of data points, geometrically spaced and optimally placed. The choice (3.4) for the weights w_n is better than the choice (3.19).

The experiments that provided the motivation for this work are in the field of photon correlation spectroscopy for the determination of molecular size distributions, and the results obtained will aid the design and operation of such experiments and photon correlation instruments. Similar Laplace inversion problems occur, however, in other areas, for example, in the measurement of fluorescent decay, in sedimentation equilibrium and in other areas of relaxation kinetics; the present work will apply equally well to the design of experiments for optimum data processing in these fields and should allow improvements in the efficiency of collection and reduction of data to be made.

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References

Bertero, M., Boccacci, P. & Pike, E. R. 1982 *Proc. R. Soc. Lond.* A **383**, 15.
Gulari, Es., Gulari, Er., Tsunashima, Y. & Chu, B. 1979 *J. chem. Phys.* **70**, 3965.
McWhirter, J. G. 1980 *Optica Acta* **27**, 83.
McWhirter, J. G. 1981 *Optica Acta* **28**, 1453.
McWhirter, J. G. & Pike, E. R. 1978 *J. Phys.* A **11**, 1729.
Miller, G. F. 1974 *Numerical solution of integral equations* (ed. L. M. Delves & J. Walsh), p. 175. Oxford: Clarendon Press.
Ostrowsky, N., Sornette, D., Parker, P. & Pike, E. R. 1981 *Optica Acta* **28**, 1059.
Pike, E. R., Watson, D. & McNeil-Watson, F. 1982 *Proc. 1st Conference American Aerosol Association, Santa Monica* (ed. B. Dahneke). New York: Wiley.
Ruhe, A. 1980 *SIAM Jl Sci. Stat. Comut.* **1**, 481.
Provencher, S. W. 1979 *Makromolek. Chem.* **180**, 201.