General Moment Methods for a Class of Nonlinear Models

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ABSTRACT

This paper deals with the following problems: (i) the development of a general theory which incorporates several methods as special cases; (ii) the applicability of moment methods to a class of non-linear problems, (iii) the specification of the class of admissible weighting functions; (iv) the estimation of the number of parameters; (v) the elimination of the cut-off error in the analysis of fluorescence decay data.

1. Introduction

There are several methods in the literature which are commonly referred to as the method of moments. For example there is a classical method of moments involved in the computation of the eigenvalues of an integral operator [1]. Another method of moments [2] which is widely used is the analysis of fluorescence decay experimental data, deals with computing the decay rates $\lambda_i < 0$, $i = 1, 2, \dots, N$ and corresponding amplitudes $\alpha_i > 0$, $i = 1, 2, \dots, N$, in the model:

$$x(t) = \sum_{i=1}^N \alpha_i \exp(\lambda_i t), \quad 0 \leq t < \infty \quad (1.1)$$

$$F(t) = \int_0^t e(s)x(t-s)ds, \quad 0 \leq t < \infty \quad (1.2)$$

from discrete data $F(t_i)$, $e(t_i)$ ($i = 1, 2, \dots, q$). This method has been developed by I. Isenberg and his colleagues. (See [2] - [7]). We will refer to it as IMOM (Isenberg method of moments). A third method of moments, which we will call LAM (linear algebraic method) was introduced for nonlinear models of the form

$$x(t) = \sum_{i=1}^N \alpha_i \phi(t; \lambda_i), \quad a \leq t \leq b. \quad (1.3)$$

where the parameters λ_i , α_i ($i = 1, 2, \dots, N$) are computed from discrete observations of $x(t)$. Nonlinear models such as (1.3) and the fluorescence decay model (1.1) - (1.2) are usually considered from the point of view of nonlinear least squares ([8] - [11]). In fluorescence decay experiments, where both IMOM and nonlinear least squares are both among the standard methods, IMOM has been found to have several advantages. For a comparison of methods see [2], [10]

and especially [12, Section 6]. In general moment methods do not require an initial approximations (starting values) and require less computer time [12]. Moreover moment methods offer a direct means for computing the number of unknown parameters (i.e., the integer N in Equations (1.1) and (1.3)) when this is not already specified by the model [13].

The objective of this paper is twofold. First, using linear operator theory, a method of moments is developed which is general enough to include all of the moment methods discussed above. We call this method GMOM (general method of moments). Second, GMOM is used to derive a new method which we call DCM (direct computational method). DCM, applied to the model (1.1) - (1.2) alleviates some long standing difficulties arising in IMOM: the cut-off error and lack of flexibility in weighting the data. Numerical examples are presented in Section 8.

2. Algebraic principles underlying moment methods

Consider a nontrivial sequence of real numbers s_k ($k = 0, 1, \dots, m$) which may be represented in terms of real numbers a_i, λ_i ($i = 1, 2, \dots, n$) by Equation (2.1).

$$s_k = \sum_{i=1}^n a_i \lambda_i^k, \quad (k = 0, 1, \dots, m). \quad (2.1)$$

We will refer to such a sequence (2.1) as a moment sequence since $s_k = \int_{-\infty}^{\infty} \lambda^k dm$ where $m(\lambda)$ is a step function which jumps a_i at $\lambda = \lambda_i$ ($i = 1, 2, \dots, n$). Moment methods are ultimately based on estimating a moment sequence from numerical data via the equations which comprise

the model (a_i and λ_i are not known in advance). It is always possible to express s_k ($k = 0, 1, \dots, m$) in the form $s_k = \sum_{i=1}^N b_i v_i^k$ where $b_i \neq 0$ and the v_i are distinct numbers forming a subset of $\{\lambda_i\}$. We may assume (by relabeling λ_i) that $v_i = \lambda_i$ ($i = 1, 2, \dots, N$). Thus

$$s_k = \sum_{i=1}^N b_i \lambda_i^k \quad (2.2)$$

where

$$\lambda_i \neq \lambda_j \text{ (} i \neq j \text{) and } b_i \neq 0 \text{ (} i = 1, 2, \dots, N \text{).} \quad (2.3)$$

The representation (2.2) where the parameters are subject to condition (2.3) is called canonical (see [14, p.3]). The integer N , called the number of components, may not be known in advance, however, we usually assume that m is sufficiently large so that

$$m \geq 2N + 1. \quad (2.4)$$

A moment sequence $\{s_k\}$ is said to be positive [14] if in the canonical representation (2.2) we have

$$b_i > 0 \text{ (} i = 1, 2, \dots, N \text{)} \quad (2.5)$$

(positivity is useful for numerical estimation of N).

The numerical procedure for estimating the number of components N and the parameters $\{\lambda_i\}$ and $\{b_i\}$ is based on the following results.

Proposition 1

Let $\{s_k\}$ be a moment sequence, let $\{G_k\}$ denote the Hankel matrices

$$G_k = \begin{pmatrix} s_0 & s_1 & \dots & s_{k-1} \\ s_1 & s_2 & \dots & s_k \\ \cdot & \cdot & & \\ \cdot & \cdot & & \\ \cdot & \cdot & & \\ s_{k-1} & s_k & \dots & s_{2k-2} \end{pmatrix} \quad (2.6)$$

and let $|G_k|$ denote the determinant of G_k . Then the following statements are true:

1.

$$|G_N| \neq 0 \quad (2.7)$$

$$|G_k| = 0 \text{ if } k > N \quad (2.8)$$

In the event that $\{s_k\}$ is positive we have further:

$$|G_k| > 0 \text{ if } k \leq N. \quad (2.9)$$

2. The numbers λ_k are the roots of a polynomial

$$P_N(\lambda) = \lambda^N + c_{N-1}\lambda^{N-1} + \dots + c_0 \quad (2.10)$$

where the coefficients c_i are given by

$$\begin{pmatrix} c_0 \\ c_1 \\ \cdot \\ \cdot \\ \cdot \\ c_{N-1} \end{pmatrix} = -G_N^{-1} \begin{pmatrix} s_N \\ s_{N+1} \\ \cdot \\ \cdot \\ \cdot \\ s_{2N-1} \end{pmatrix}. \quad (2.11)$$

3. The numbers b_i are given by

$$\begin{pmatrix} b_1 \\ b_2 \\ \cdot \\ \cdot \\ \cdot \\ b_N \end{pmatrix} = V_N^{-1} \begin{pmatrix} s_0 \\ s_1 \\ \cdot \\ \cdot \\ \cdot \\ s_{N-1} \end{pmatrix} \quad (2.12)$$

where V_N is the Vandermonde matrix

$$V_N = \begin{pmatrix} 1 & 1 & \cdot & \cdot & \cdot & 1 \\ \lambda_1 & \lambda_2 & \cdot & \cdot & \cdot & \lambda_N \\ \cdot & \cdot & & & & \\ \cdot & \cdot & & & & \\ \cdot & \cdot & & & & \\ \lambda_1^{N-1} & \lambda_2^{N-1} & & & & \lambda_N^{N-1} \end{pmatrix}. \quad (2.13)$$

Proof

(1) The matrix G_k may be factored in the form

$$G_k = V_k D_N V_k^T \quad (2.14)$$

(V_k^T is the transpose of V_k) where D_N is the $N \times N$ diagonal matrix

$$D_N = \text{diag}(b_1, b_2, \dots, b_N) \quad (2.15)$$

and V_k is the $k \times N$ Vandermonde matrix whose j th row is $(\lambda_1^{j-1}, \lambda_2^{j-1}, \dots, \lambda_N^{j-1})$. In view of condition (2.3) the rank of D_N is N while $\text{rank}(V_k) = k$ if $k \leq N$ and $\text{rank}(V_k) = N$ if $k > N$. Thus $\text{rank}(G_N) = N$ and $\text{rank}(G_k) < k$ if $k \geq N$. This proves Equations (2.7) and (2.8). Moreover

if condition (2.5) is satisfied then D_N , and hence also G_N , is positive definite and Equation (2.9) is satisfied. This concludes the proof of the first statement.

(2) Let $P_N(\lambda)$ be the monic polynomial whose roots are λ_i ($i = 1, 2, \dots, N$). For each natural number k we have $b_i \lambda_i^k P_N(\lambda_i) = 0$. Summing over $i = 1, 2, \dots, N$ yields $s_{N+k} + \sum_{i=0}^{N-1} c_i s_{i+k-1} = 0$, $k = 0, 1, \dots, N-1$ where the c_i are the coefficients of $p_N(\lambda)$. This system of equations is equivalent to Equation (2.11).

(3) The system (2.12) is equivalent to Equations (2.2) for $k = 0, 1, \dots, N-1$. This completes the proof.

Proposition 1 is essentially an algorithm for computing N , $\{\lambda_i\}$ and $\{b_i\}$ from the moment sequence. To find N the sequence of determinants $|G_k|$ is computed until one observes a string of zeros. Observe that to find at least one zero we must have condition (2.4). Numerical difficulties do arise in identifying zero and therefore appropriate scaling (see [13]) is essential. It was found in [13] that, in the case of positive moment sequences, a negative computed value indicates the number of components i.e., $|G_{k+1}| < 0$ indicates $k = N$.

Once N is assumed known, the next step is to estimate the λ_i ($i = 1, 2, \dots, N$) by calculating the roots of $P_N(\lambda)$. Finally, the sequence $\{b_i\}$ is found from Equation (2.12).

3. GMOM (general method of moments)

In the previous section we saw that a simple algebraic procedure could be utilized to estimate parameters from a moment sequence. This

section deals with classifying, in general terms, the class of problems for which a moment sequence may be defined. For this purpose we work in a Hilbert space H . Let $\phi(\lambda)$ be a function from the real numbers into H . We consider a model of the form

$$x = \sum_{i=1}^n \alpha_i \phi(\lambda_i), \quad \alpha_i \neq 0 \quad (i = 1, 2, \dots, n) \quad (3.1)$$

$$f = Ex. \quad (3.2)$$

We do not assume that $x, n, \{\alpha_i\}, \{\lambda_i\}$ are known in advance but we do require that enough information is provided to estimate the numbers s_k defined below. Here E is a linear operator mapping its domain, $\text{Dom}(E) (\subset H)$, into H . Observe that the model (3.1) - (3.2) is an obvious generalization of (1.1) - (1.2).

Since Equation (3.1) is nonlinear in λ_i ($i = 1, 2, \dots, n$) we have little chance of applying a moment method (which is essentially linear) unless we can associate $\{\lambda_i\}$ with a linear operator. Therefore, we assume that there is a linear operator $L: \text{Dom}(L) \rightarrow H$ ($\text{Dom}(L) \subset H$) such that

$$L\phi_i = \lambda_i \phi_i, \quad (i = 1, 2, \dots, n) \quad (3.3)$$

where (we will henceforth write)

$$\phi_i = \phi(\lambda_i). \quad (3.4)$$

It is assumed that ϕ_i ($i = 1, 2, \dots, n$) are linearly independent in H .

Let N be the number of distinct λ_i appearing in Equation (3.1) (if L does not have eigenvalues of multiplicities > 1 then $N = n$) and let m be an integer such that $m \geq 2N-1$. We say that an element h is

admissible if it satisfies the following conditions.

$$A. \quad h \in \text{Dom}[(L^*)^m E^*] \quad (3.5)$$

(* denotes adjoint).

B. There is a sequence of linear operators A_k such that $h \in \text{Dom}(E^* A_k)$ and

$$E^* A_k h = (L^*)^k E^* h \quad (k = 0, 1, \dots, m) \quad (3.6)$$

(A_0 may be taken to be the identity in H).

C. The elements

$u_k = (L^*)^k E^* h \quad (k = 0, 1, \dots, N-1)$ are linearly independent (3.7) as members of S^* where $S = \text{span}(x, Lx, \dots, L^{N-1}x)$.

Theorem 1

Let h be admissible. Then the sequence s_k defined by

$$s_k = \langle f, A_k h \rangle \quad (k = 0, 1, \dots, m) \quad (3.8)$$

is a moment sequence ($\langle \cdot, \cdot \rangle$ denotes the inner product in H). In fact,

$$s_k = \sum_{i=1}^n a_i \lambda_i^k \quad (k = 0, 1, \dots, n). \quad (3.9)$$

where

$$a_i = \alpha_i \langle \phi_i, E^* h \rangle \quad (i = 1, 2, \dots, n) \quad (3.10)$$

Moreover, the number of components associated with the moment sequence $\{s_k\}$ is N (recall N is the number of distinct eigenvalues associated with the n eigenvectors ϕ_i).

Proof

Under the substitution from Equation (3.2), Equation (3.8) becomes

$s_k = \langle E x, A_k h \rangle = \langle x, E^* A_k h \rangle$. In view of condition (3.6),

$$s_k = \langle x, (L^*)^k E^* h \rangle. \quad (3.13)$$

From Equations (3.1) and (3.4), $s_k = \sum_{i=1}^n \alpha_i \langle \phi_i, (L^*)^k E^* h \rangle$
 $= \sum_{i=1}^n \alpha_i \langle L^k \phi_i, E^* h \rangle$. With the application of Equation (3.3) we obtain
the representation (3.8) - (3.9).

Observe from Equation (3.13) that

$$s_{i+j} = \langle L^i x, u_j \rangle. \quad (3.14)$$

From Equations (3.1) and (3.3), and from the fact that the ϕ_i
($i = 1, 2, \dots, n$) are linearly independent, it can be seen that the set
 $L^i x$ ($i = 0, 1, \dots, N-1$) is a linear independent in H . Since the sequence
 u_j ($j = 0, 1, \dots, N-1$) is also linear independent by assumption, it
follows from the Gramian structure of G_N (defined in Equation (2.6))
in terms of $\{L^i x\}$ and $\{u_j\}$ (see Equation (3.14)) that the matrix G_N
is nonsingular. Hence, from Proposition 1 (see Equation (2.8)) the
number of components $\geq N$. Since there are only N distinct λ_i appearing
in Equation (3.8), the number of components is precisely N . This com-
pletes the proof.

Remark 1

The theorem requires that $m \geq 2N - 1$ for otherwise G_N could not
be constructed. If we wish to detect N by means of Equation (2.8)
then we need to construct G_{N+1} and this requires $m \geq 2N + 1$.

Remark 2

Observe that each admissible h gives rise to a moment sequence from
which one estimates, via Proposition 1, the set of distinct λ_i

appearing in Equation (3.1). If N is known in advance it suffices to choose $m = 2N - 1$. In the event that $n = N$, which is necessarily true when the eigenvalues of L are simple, then a_i and, hence also α_i , ($i = 1, 2, \dots, n$) are obtained from Equations (2.12) and (3.10) (note that $a_i = b_i$ ($i = 1, 2, \dots, N$) in this case).

Remark 3

Suppose L has simple eigenvalues then condition C is equivalent to the condition:

$$\langle \phi_i, E^*h \rangle \neq 0, \quad i = 1, 2, \dots, N. \quad (3.15)$$

In fact it follows directly from the proof that s_k has the representation $s_k = \sum_{i=1}^N \alpha_i \langle \phi_i, E^*h \rangle \lambda_i^k$ in this case.

In each of the models presented below H is a space of square integrable functions and the s_k ($k = 0, 1, \dots, m$) defined in Equation (3.8) turn out to be integrals. Different choices of admissible functions correspond to different weightings of the data. Thus h may be viewed as a weighting function.

4. Eigenvalues of integral operator

The Ritz method [1] is a procedure for estimating the eigenvalues of a symmetric integral operator K defined on Hilbert space $L_2[a, b]$. To employ the Ritz method a set of linear independent coordinate functions u_i ($i = 1, 2, \dots$) are chosen. Let P be the orthogonal projection onto the subspace H which is spanned by u_i ($i = 1, 2, \dots, n$). Then the linear operator L , the restriction of PK to H , is a symmetric

operator mapping H into itself. The eigenvalues of L are approximations of the eigenvalues of K . The classical method known as "the method of moments" is a special case of the Ritz method in which the coordinate functions have the form

$$u_k = L^k h, \quad (k = 0, 1, \dots, n-1) \quad (4.1)$$

where h is a particular function in H for which the sequence u_k ($k = 0, 1, \dots, n-1$) is linear independent.

This method of moments may be put into the general framework of the previous section if we equate

$$x = f = h \quad (4.2)$$

and set

$$E = I \text{ (the identity operator in } H\text{)}. \quad (4.3)$$

Let ϕ_i ($i = 1, 2, \dots, n$) be the eigenvectors of L in H corresponding to eigenvalues λ_i ($i = 1, 2, \dots, n$). Also set $\phi(\lambda) = \sum_{i=1}^n \delta(\lambda - \lambda_i) \phi_i$ (δ is the Kronecker delta function). The condition that u_k ($k = 0, 1, \dots, n-1$) is linear independent implies condition (3.7) (since $N \leq n$) and it also guarantees, that in the representation

$$h = \sum_{i=1}^n \alpha_i \phi_i, \quad (4.4)$$

the coefficients $\alpha_i \neq 0$ ($i = 1, 2, \dots, n$). Choosing

$$A_k = L^k, \quad k = 0, 1, \dots, m, \quad (4.5)$$

and noting that $L^* = L$, we see that all the conditions needed for the application of GMOM are satisfied. In this particular case, since n

and x is known in advance, a complete set of moments may be obtained from Equation (3.14).

$$s_{i+j} = \langle L^i h, L^j h \rangle \quad (i, j = 0, 1, \dots, n). \quad (4.6)$$

We know from Theorem 1 that s_k has the representation (3.9) where N is the number of distinct eigenvalues, however the substitution of (4.4) into (4.6) leads to representation (2.2) with $b_i > 0$ ($i = 1, 2, \dots, N$). The fact that $\{s_k\}$ is a positive moment sequence enhances the computation of N as discussed in Section 2.

To summarize, the classical method of moments for computing eigenvalues is a special case of GMOM. A function h is admissible if the functions u_k , defined in Equation (4.1), are linearly independent.

5. LAM (linear algebraic method)

We consider a computational algorithm developed in [13] to estimate N , λ_i , α_i ($i = 1, 2, \dots, N$) such that

$$x(t) = \sum_{i=1}^N \alpha_i \exp(\lambda_i t), \quad 0 \leq t \leq T \quad (5.1)$$

from discrete data for $x(t)$. The model requires that

$$\alpha_i > 0 \quad (i = 1, 2, \dots, N)$$

and the λ_i ($i = 1, 2, \dots, N$) are considered to be distinct.

To derive LAM from GMOM we set $H = L_2[0, T]$, $\phi(\lambda) = \exp(\lambda(\cdot))$, $E = I$, $f = x$ and we define L as follows:

$$Lx = x' \quad (\text{the derivative of } x), \quad (5.2)$$

$$\text{Dom}(L) = \{x | x' \in H\} \quad (5.3)$$

This implies that

$$L^*x = -x', \quad (5.4)$$

$$\text{Dom } (L^*) = \{x | x' \in H, x(0) = x(T) = 0\} \quad (5.5)$$

In order to satisfy condition B (i.e., Equation (3.6)) we take

$$A_k = (-L)^k, \quad k = 0, 1, \dots, m \quad (5.6)$$

condition (3.5) requires that an admissible function h be m times differentiable in H and

$$h^{(k)}(0) = h^{(k)}(T) = 0 \quad (k = 0, 1, \dots, m-1). \quad (5.7)$$

Observe that the eigenvalues of L are simple. In view of Remark 3 (below Theorem 1), Condition C is satisfied if $\langle \phi_i, h \rangle \neq 0$ $i = 1, 2, \dots, N$.

The moment sequence (see Equation (3.8)) is defined by

$$s_k = (-1)^k \langle x, h^{(k)}(t) \rangle \quad (k = 0, 1, \dots, m) \quad (5.8)$$

and s_k has the representation (see Equations (3.9) and (3.10))

$$s_k = \sum_{i=1}^N \alpha_i \langle \phi_i, h \rangle \lambda_i^k$$

where $\langle \phi_i, h \rangle = \int_0^T \exp(\lambda_i t) h(t) dt$. If $h(t) \geq 0$ in $[0, T]$ (and not identically zero) then $a_i = \alpha_i \langle \phi_i, h \rangle > 0$ ($i = 1, 2, \dots, N$) so that s_k ($k = 0, 1, \dots, m$) is a positive moment sequence. An obvious choice for h is

$$h(t) = t^m (T - t)^m. \quad (5.9)$$

In conclusion, LAM is a special case of GMOM. A sufficient

condition for h to be admissible is that h is m times differentiable, satisfies conditions (5.7) and h is nonnegative (but not identically zero).

6. IMOM (Isenberg method of moments)

IMOM is a procedure for estimating the "decay rates" λ_i ($i = 1, 2, \dots, N$) and the corresponding amplitudes α_i ($i = 1, 2, \dots, N$) subject to the "fluorescence decay equations"

$$x(t) = \sum_{i=1}^n \alpha_i \exp(\lambda_i t) \quad (t \geq 0) \quad (6.1)$$

$$f(t) = \int_0^t e(t-s) x(s) ds \quad (t \geq 0). \quad (6.2)$$

The function $e(t)$ is positive, continuous and has compact support in $(0, \infty)$. Typically about 500 equally spaced observations for $f(t)$ and $e(t)$ are available. The λ_i ($i = 1, 2, \dots, N$) are distinct and it is also assumed in the model that

$$\alpha_i > 0, \quad \lambda_i < 0 \quad (i = 1, 2, \dots, N). \quad (6.3)$$

We will derive IMOM (see [2]) from GMOM. For this purpose let $H = L_2 [0, \infty]$, $\phi(\lambda) = \exp(\lambda(\cdot))$,

$$Ex(t) = \int_0^t e(t-s) x(s) ds \quad (\text{Dom}(E) = H), \quad (6.4)$$

$$Lx(t) = x'(t), \quad \text{Dom}(L) = \{x \mid x' \in H, x(\infty) = 0\}. \quad (6.5)$$

This implies

$$E^*x(t) = \int_0^\infty e(s) x(s+t) ds \quad (\text{Dom}(E^*) = H) \quad (6.6)$$

$$L^*x(t) = -x'(t), \quad \text{Dom}(L^*) = \{x \mid x' \in H, x(0) = 0\}. \quad (6.7)$$

To satisfy condition (3.6) we define

$$A_k x(t) = (-1)^k x^{(k)}(t), \quad \text{Dom}(A_k) = \{x \mid x^{(k)} \in H\} \quad (6.8)$$

($k = 0, 1, \dots, m$). Condition (3.5) requires that an admissible function

h be m times differentiable in H and

$$w^{(k)}(0) = 0 \quad (k = 0, 1, \dots, m-1) \quad (6.9)$$

where

$$w(t) = E^*h(t) \quad (6.10)$$

Here, as in the previous model, the eigenvalues of L are simple. Thus condition C is reduced to condition (3.15) which is clearly satisfied whenever $w \geq 0$ (but not identically zero).

If h is chosen such that

$$w(t) = t^m/m! \quad (6.11)$$

(see Equations (6.14) through (6.17)) then it follows from Equations (3.9) and (3.10) that

$$s_k = \sum_{i=1}^n \alpha_i \left[\int_0^\infty \exp(\lambda_i t) w(t) dt \right] \lambda_i^k = \sum_{i=1}^n \alpha_i (-1)^{n+1} \lambda_i^{(k-m-1)}.$$

If we set $g_k = (-1)^{m-k} s_{m-k}$ and $\tau_i = -1/\lambda_i$, then

$$g_k = \sum_{i=1}^n \alpha_i \tau_i^k \quad (k = 0, 1, \dots, m) \quad (6.12)$$

The moments sequence $\{g_k\}$ is obtained in IMOM. We have shown that these numbers are also obtained as a special case of GMOM provided h is chosen so that w has the form (6.11). In order to construct this particular h we set

$$m_k = \int_0^\infty t^k e(t) dt, \quad k = 0, 1, \dots, m \quad (6.13)$$

and define recursively

$$\phi_0 = 1/m_0 \quad (6.14)$$

$$\phi_k = \frac{t^k}{m_0} - \frac{1}{m_0} \sum_{i=1}^k \binom{k}{i} m_i \phi_{k-i} . \quad (6.15)$$

It is not difficult to see that

$$E^* \phi_k = t^k \quad (6.16)$$

so that the appropriate h is given by

$$h(t) = \phi_m(t)/m! \quad (6.17)$$

The method presented here is essentially the same as IMOM although the order in which calculations are performed is different. One reason for presenting IMOM from a more general point of view is to show that there exist many choices of weighting functions. In fact it is sufficient that $w = E^*h \geq 0$ (but not identically zero) and satisfy condition (6.9).

The particular choice (6.17) has the disadvantage that the estimation of the moment sequence requires the computation of integrals of the form

$$\int_0^\infty t^k f(t) dt \quad (k = 0, 1, \dots, m). \quad (6.18)$$

Since data for $f(t)$ is given in a finite interval $[0, T]$ one incurs the so called cut-off error. IMOM attempts to minimize the effects of cut-off error by a combination of exponential depression and extrapolation (see [2] for details). However one can completely avoid the error by working in the space $L_2 [0, T]$ as we show in Section 7.

In IMOM the moment sequence which is obtained is positive. However, IMOM makes no attempt to compute N .

7. DCM (direct computation method)

In this section we consider a model of the form

$$x(t) = \sum_{i=1}^N \alpha_i \exp(\lambda_i t) \quad (0 \leq t \leq T) \quad (7.1)$$

$$f(t) = \int_0^t e(t-s) x(s) ds \quad (0 \leq t \leq T) \quad (7.2)$$

The $\{\lambda_i\}$ are distinct and the $\{\alpha_i\}$ are nonzero. For the sake of generality, condition (6.3) will not be assumed. The function $e(t)$ is continuous and not identically zero. The derivation of DCM is similar to the derivation of IMOM although there are important differences. We will only point out these differences. The space H is now $L_2 [0, T]$. With this definition of H understood, the operator E is defined in Equation (6.4). However, now, instead of Equation (6.6), we have

$$E^* x(t) = \int_0^{T-t} e(s) x(s+t) ds \quad (\text{Dom } (E^*) = H) \quad (7.3)$$

Since the interval is now finite Equations (6.5) and (6.7) are replaced by

$$Lx(t) = x'(t), \quad \text{Dom } (L) = \{x \mid x' \in H\} \quad (7.4)$$

$$L^*x(t) = -x'(t), \quad \text{Dom } (L) = \{x \mid x' \in H, x(0) = x(T) = 0\} \quad (7.5)$$

The operators A_k are defined as in Equation (6.8). In place of Equation (6.9) we now have

$$w^{(k)}(0) = w^{(k)}(T) = 0 \quad (k = 0, 1, \dots, m-1) \quad (7.6)$$

where w is given in terms of h by Equation (6.10). Once again we

may replace condition C with condition (3.15). Thus h is an admissible function if it is m times differentiable in H and $w (=E^*h)$ satisfies conditions (7.6) and

$$\int_0^T \exp(\lambda_i t) w(t) dt \neq 0, \quad i = 1, 2, \dots, N \quad (7.7)$$

It is not difficult to see that conditions (7.6) are satisfied if

$$h^{(k)}(T) = 0, \quad \langle e, h^{(k)} \rangle = 0 \quad (k = 0, 1, \dots, m-1) \quad (7.8)$$

Thus it is natural to choose h in the form

$$h(t) = (T - t)^m q(t) \quad (7.9)$$

where

$$q(t) = t^m + d_{m-1} t^{m-1} + \dots + d_0. \quad (7.10)$$

Substitution of Equations (7.9) and (7.10) into the second set of Equations in (7.8) gives rise to an $m \times m$ system of equations for $d_0, d_1, \dots, d_{m-1}$, any solution of which yields a function satisfying condition (7.6).

An alternative method for computing h in the form (7.9) - (7.10) which does not require the solving of a system of equation, assumes that $m_0 = \int_0^T e(t) dt \neq 0$. We compute the integrals $m_k = \int_0^T t^k e(t) dt$ ($k = 0, 1, \dots, m$) and define the functions $\phi_k(t)$ ($k = 0, 1, \dots, m$) as in Equations (6.14) and (6.15). We then define recursively

$$\psi_{1,j} = \phi_j \quad (j = 0, 1, \dots, m) \quad (7.11)$$

$$\psi_{i+1,j} = A_{i,j} \psi_{i,j} - B_{i,j} \psi_{i,j+1} \quad (7.12)$$

where $A_{i,j}$ and $B_{i,j}$ are constants defined by

$$A_{i,j} = \psi_{i,j+1}^{(i-1)}(T), \quad B_{i,j} = \psi_{i,j}^{(i-1)}(T). \quad (7.13)$$

We then set

$$h = \psi_{m,m-1}. \quad (7.14)$$

If h is given by Equation (7.14) then $w = E^*h$ will satisfy conditions (7.6) but we can not insure that it also satisfies condition (7.7) for all possible λ_i but, in practice, this does not appear to be a problem.

Once an admissible function h is obtained the moment sequence is given by (see Equation (3.8))

$$s_k = (-1)^k \int_0^T f(t) h^{(k)}(t) dt \quad (k = 0, 1, \dots, m). \quad (7.15)$$

The model parameters N, λ_i, α_i ($i = 1, 2, \dots, N$) are estimated in the manner prescribed in Sections 2 and 3.

Numerical examples and a comparison of DCM with IMOM are presented below. From these examples we know that, when the weighting function h is chosen in the manner discussed above, DCM tends to demonstrate greater accuracy than IMOM. However, the problem of finding an optimal weighting function has not been resolved.

8. Numerical Examples

Numerical examples for LAM and IMOM can be found in [13], [2], [3], [6], [7] and [12]. In this section we will illustrate the performance of the proposed DCM with several numerical examples using computer simulated data. For comparison, we also present the results obtained by analysis of the data by IMOM. All computations were performed in double precision on the DEC System 10 in time-sharing conversational fashion (located at The University of Texas Health Science Center at Dallas, Texas).

The model is of the form

$$\begin{aligned}x(t) &= \sum_{i=1}^N \alpha_i \exp(-t/\tau_i) \quad (0 \leq t \leq T), \\f(t) &= \int_0^t e(t-s) x(s) ds \quad (0 \leq t \leq T),\end{aligned}$$

and let $e(t)$ be chosen so that

$$\begin{aligned}e(t) &= \beta \sin \pi t / \hat{T} \quad (0 \leq t \leq \hat{T}) \\&= 0 \quad (\hat{T} < t).\end{aligned}$$

The data sets for $f(t)$ and $e(t)$ were generated as described in [12]. The noise which is distributed in a Gaussian fashion is also added to $f(t)$. Many different analyses were carried out for the purpose of comparing the results of various combinations of parameters (N , α_i , τ_i , β , $\hat{T}$ and T). The admissible functions are chosen as Equation (7.13) for all analyses in this section. The experimental data sets were created by choosing $\beta = 10^5$, $\hat{T} = 30$, $T = 500$, $N = 1, 2, 3$ and different combinations of α_i and τ_i for the analysis

presented in Table I and II. Table I gives a comparison of the expected parameters and the calculated results obtained by analysis of the data by DCM for different sampling intervals. For comparison of the accuracy of the estimated parameters with the expected values, the relative errors (in percent) of the estimated parameters are presented in Table II, along with the results obtained by IMOM. The cpu times (in second) are also presented. The relative errors of the estimated parameters of a typical two component case ($\alpha_1 = 1$, $\tau_1 = 15$, $\alpha_2 = 1$, $\tau_2 = 30$) with varying parameters β and $\hat{T}$ are given in Table III. It should be pointed out that all integrals in the computation were evaluated using Simpson's Rule on both DCM and IMOM. Also exponential depression (this is related to the cut-off error), moment index displacement (see [2] for an explanation of these parameters) and scaling the calculated moments were employed for IMOM in the computation.

Study of Table II and III reveals that DCM always provides nice results for the sampling interval $[0, 500]$. DCM also provides acceptable results even in the cases of smaller sampling intervals. In most cases the larger the sampling interval, the greater the accuracy with only a few exceptions. When β or $\hat{T}$ is larger, the $f(t)$ is larger. Consequently, the signal-to-noise ratio will be better yielding more accurate results as illustrated in Table III. It also should be pointed out that for either greater intervals or for larger N , the number of calculations will be increased, thereby increasing the cpu time requirements. Though the cpu time

requirements in the DCM are more than for IMOM, the error committed by the former method is far less than the latter in most cases. An important point to note is that IMOM may not work satisfactorily in those cases where the exponential depression parameters are not given correctly. One of the major difficulties in the implementation of IMOM is not knowing the appropriate choice of the exponential depression parameter. In DCM, where there is no cut-off error, there is no need for exponential depression and the problem does not arise. The conclusion we can draw from these experiments (and from other experiments we have performed) is that: (i) from the point of view of computation time, IMOM is a better method than DCM, (ii) from the point of view of accuracy, DCM is superior to IMOM. Moreover, for the fluorescence decay problem at hand, DCM is also efficient. A detailed description of the application of DCM in the analysis of fluorescence decay will appear elsewhere.

TABLE I

Comparison of the Estimated Parameters with the Expected Values by DCM

		Expected Values	[0,100]	[0,200]	Interval [0,300]	[0,400]	[0,500]
1	α	1.00	1.004	1.003	1.003	1.003	1.002
	τ	15.00	14.989	14.992	14.993	14.993	14.993
2	α	0.15	0.150	0.150	0.150	0.150	0.150
	τ	18.40	18.392	18.398	18.399	18.400	18.400
3	α_1	1.00	1.012	1.011	1.012	1.013	1.013
	τ_1	5.00	5.087	5.009	4.993	4.984	4.978
	α_2	1.00	0.991	0.998	0.998	0.999	0.999
	τ_2	30.00	30.162	30.018	30.008	30.004	30.002
4	α_1	1.00	1.270	1.048	1.023	1.016	1.013
	τ_1	12.50	13.358	12.663	12.569	12.540	12.529
	α_2	0.50	0.228	0.451	0.477	0.484	0.487
	τ_2	19.50	23.329	19.872	19.664	19.611	19.591
5	α_1	0.10		0.109	0.103	0.101	0.101
	τ_1	5.00		4.560	4.904	4.999	4.994
	α_2	0.017		0.016	0.017	0.017	0.017
	τ_2	30.00		26.418	29.768	30.310	30.117
	α_3	0.0084		0.0110	0.0086	0.0083	0.0084
	τ_3	60.00		55.885	59.607	60.043	59.907
6	α_1	1.00		1.032	1.040	1.031	1.026
	τ_1	7.50		7.429	7.682	7.653	7.632
	α_2	0.42		0.425	0.395	0.397	0.399
	τ_2	18.40		19.120	19.477	19.127	18.983
	α_3	0.18		0.161	0.164	0.171	0.174
	τ_3	35.80		36.671	36.377	36.095	36.012

TABLE II

Comparison of the Accuracy of the Parameters as Estimated by Different Methods

(Relative error in percentage, cpu in seconds, expected values given in Table I)

		IMOM			DCM		
		[0,500]	[0,100]	[0,200]	[0,300]	[0,400]	[0,500]
1	$\Delta\alpha$	0.24	0.40	0.28	0.26	0.25	0.24
	$\Delta\tau$	0.11	0.07	0.05	0.05	0.05	0.05
	cpu	3.34	1.33	4.03	8.25	14.02	21.35
2	$\Delta\alpha$	0.21	0.21	0.10	0.08	0.07	0.06
	$\Delta\tau$	0.11	0.05	0.01	0.00	0.00	0.00
	cpu	3.31	1.33	4.02	8.27	14.03	21.36
3	$\Delta\alpha_1$	6.64	1.20	1.13	1.22	1.28	1.34
	$\Delta\tau_1$	1.55	1.74	0.18	0.13	0.32	0.44
	$\Delta\alpha_2$	0.76	0.88	0.23	0.17	0.13	0.12
	$\Delta\tau_2$	0.02	0.54	0.06	0.03	0.01	0.01
	cpu	3.39	4.44	11.73	22.11	35.58	52.00
4	$\Delta\alpha_1$	3.42	27.00	4.86	2.28	1.56	1.28
	$\Delta\tau_1$	0.18	6.87	1.30	0.55	0.32	0.23
	$\Delta\alpha_2$	4.29	54.42	9.84	4.59	3.12	2.56
	$\Delta\tau_2$	0.32	19.64	1.90	0.84	0.57	0.46
	cpu	3.33	4.44	11.74	22.13	35.58	52.01
5	$\Delta\alpha_1$	6.07		9.02	2.73	1.05	0.71
	$\Delta\tau_1$	0.85		8.80	1.93	0.03	0.13
	$\Delta\alpha_2$	15.77		30.66	1.04	1.03	1.00
	$\Delta\tau_2$	2.44		11.94	0.77	1.03	0.39
	$\Delta\alpha_3$	11.96		7.15	2.44	0.98	0.14
	$\Delta\tau_3$	1.53		6.86	0.65	0.07	0.15
	cpu	3.39		26.78	46.98	1:11.78	1:41.53
6	$\Delta\alpha_1$	9.66		3.17	4.03	3.07	2.63
	$\Delta\tau_1$	9.78		0.94	2.43	2.04	1.76
	$\Delta\alpha_2$	23.87		0.47	6.00	5.53	4.98
	$\Delta\tau_2$	6.41		3.91	5.86	3.95	3.17
	$\Delta\alpha_3$	0.61		10.35	8.64	4.83	3.57
	$\Delta\tau_3$	0.78		2.43	1.61	0.83	0.59
	cpu	6.01		26.83	46.96	1:11.85	1:41.08

TABLE III

Comparison of the Accuracy of the Parameters as Estimated by Different β and $\hat{T}$
(Relative error in percentage)

		IMOM		DCM			
		[0,500]	[0,100]	[0,200]	[0,300]	[0,400]	[0,500]
1	$\beta=10^4$ $\hat{T}=10$	$\Delta\alpha_1$	11.33	53.38	9.13	3.69	2.28
		$\Delta\tau_1$	5.38	22.57	6.95	4.33	3.53
		$\Delta\alpha_2$	6.02	57.60	12.64	6.97	5.46
		$\Delta\tau_2$	0.99	32.08	3.24	1.49	1.08
2	$\beta=10^5$ $\hat{T}=10$	$\Delta\alpha_1$	6.27	49.08	8.07	3.31	1.80
		$\Delta\tau_1$	0.70	20.84	6.65	4.31	3.44
		$\Delta\alpha_2$	1.04	52.99	11.45	6.47	4.85
		$\Delta\tau_2$	0.10	27.38	2.78	1.27	0.84
3	$\beta=10^6$ $\hat{T}=10$	$\Delta\alpha_1$	6.71	45.45	7.26	3.03	1.71
		$\Delta\tau_1$	0.17	19.49	6.20	4.12	3.36
		$\Delta\alpha_2$	0.46	49.26	10.56	6.14	4.72
		$\Delta\tau_2$	0.01	24.02	2.52	1.19	0.82
4	$\beta=10^4$ $\hat{T}=30$	$\Delta\alpha_1$	4.06	23.83	2.04	0.60	0.39
		$\Delta\tau_1$	2.48	9.09	0.96	0.24	0.13
		$\Delta\alpha_2$	3.41	24.45	2.32	0.82	0.59
		$\Delta\tau_2$	0.45	9.84	0.67	0.23	0.17
5	$\beta=10^5$ $\hat{T}=30$	$\Delta\alpha_1$	1.42	18.23	1.95	0.85	0.49
		$\Delta\tau_1$	1.26	6.91	1.04	0.47	0.25
		$\Delta\alpha_2$	0.54	18.56	2.11	0.95	0.57
		$\Delta\tau_2$	0.14	7.17	0.55	0.22	0.12
6	$\beta=10^6$ $\hat{T}=30$	$\Delta\alpha_1$	1.91	17.23	1.58	0.61	0.35
		$\Delta\tau_1$	0.44	6.53	0.84	0.34	0.19
		$\Delta\alpha_2$	0.27	17.59	1.76	0.74	0.47
		$\Delta\tau_2$	0.05	6.75	0.45	0.16	0.09

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