Application of tools of numerical analysis to a multiexponential approximation of data series

Trusov M.A., Volkov A.D.
NT–MDT Company, Zelenograd, Moscow, Russia

Fluorescence scanning confocal microscopes with pulsed laser sources include also modes for lifetime measurements. Today there is equipment available that allows to get light intensity distribution along the time scale with picosecond accuracy. Thus it's possible to obtain the time resolved curve of descending light intensity starting at the point of laser pulse trigger and finishing at the point where signal becomes substantially smaller than noise. The main problem for practical measurements here is poor signal to noise ratio. That's why the method of curve fitting by some model should be substantially tolerant to high frequency noise.

The major task, which we try to solve in this article, consists in description of basic methods and approaches which can be applied to an analysis of results of measurements for extraction of such components from the initial signal, which have an exponential time–dependence. We propose the following mathematical pattern of the investigated process:

$$y(t) = \sum_{i=1}^{n} a_i e^{-t/\tau_i} + a_0 + \varepsilon(t), \tag{1}$$

where $y(t)$ is the initial signal, a_0 , a_i , and τ_i are unknown parameters, and $\varepsilon(t)$ is a variate with zero mathematical expectation value and small dispersion describing simultaneously the prediction error and a possible noise. Parameters τ_i are called "lifetimes". Let us note that we also suppose an existence of a non–zero constant constituent a_0 in the initial signal. Because the measurement process, generally speaking, is discrete, we know the function $y(t)$ only in a finite set of points of time $\{t_i\}$. For this reason, below we will use, instead of $y(t)$, a number sequence $y_i \equiv y(t_i)$. In practice, the equidistant points of time are used, so in what follows we will choose an appropriate unit and will take $t_i = i$ (i.e. $y_i = y(i)$), $i = 0,\ldots,N$.

Up to now we told nothing about the number of exponents n. First of all we must be sure that the following inequality is realized:

$$\frac{N}{2} > n\,;$$

otherwise the number of unknown parameters will exceed the number of known data. The relation $N >> n$ is realized often, though. Let us first consider a case, when n is known and assigned beforehand. If the constant component a_0 is equal to zero, then one can calculate the lifetimes τ_i using the so–called extended Proni method [1]. Below we briefly describe this method and discuss possible improvements of it. The main idea is to construct an equation of linear prediction for the series $\{y_i\}$:

$$y_{i+p} = \sum_{j=0}^{p-1} \alpha_j y_{i+j} + e_i\,, \tag{2}$$

where α_i are unknown coefficients and e_i is a prediction error. The quantity p is called "linear prediction order". To calculate the coefficients α_i we apply the least square method using the singular value decomposition [2], leaving in the corresponding expansion of the vector $\bar{\alpha}$ only n leading terms. As for the order p, it must be in the interval

$$\frac{N}{2} > p > n\ .$$

In practice, the optimal value is $p \sim 5n - 10n$.

After calculation of the parameters α_i one should solve the characteristic equation of the power p :

$$\lambda^p = \sum_{i=0}^{p-1} \alpha_i \lambda^i , \tag{3}$$

and then select n required roots. Via these roots the lifetimes are calculated directly:

$$\tau_i = -1/\ln\lambda_i , \qquad i = 1,\ldots,n$$

The selection of the roots can be considerably simplified if one uses also the so–called "backward prediction". If we form a sequence $z_i = y_{N-i}$ and calculate for it the coefficients of linear prediction β_i, then the equation

$$\mu^p = \sum_{i=0}^{p-1} \overline{\beta}_i \mu_i , \tag{4}$$

will have roots which are inverse–conjugate with respect to the roots of equation (3): $\mu_i = 1/\overline{\lambda}_i$. The roots corresponding to veritable exponents will prove beyond the unit circumference while the other roots corresponding to noise will prove inside it, because a statistics of a random process does not change with time reversal. A comparison of complex roots of equations (3) and (4) give us a possibility to increase a precision of a prediction and to simplify a selection of required roots.

In a general case $a_0 \neq 0$ we may reduce the problem to the previous one by considering, instead of $\{y_i\}$, a series $z_i = y_i - y_{i+L}$, where L is a shift along the sequence. It is obvious that for the series $\{z_i\}$ the constant term will be equal to zero while the lifetimes will remain the same. As for quantity L, it should not be too small, because in subtracting close values y_i from each other the noise contribution becomes very essential, and it should not be too large, because an effective length of the series $\{z_i\}$ (i.e. $N - L$) is preferable to be as large as possible. We chose it as $L \sim N/2 - N/3$. Finally, when we know lifetimes τ_i, we can easy calculate the parameters a_i using an ordinary least–square method [3].

Let us now consider the situation when the number of exponential components n is not known. In this case one can use the same algorithm as above with replacement of the number n by a number n', which certainly exceeds the number of really existing exponents, and, of course, with appropriate choice of the order $p > n'$. The parameters α_i then split into two groups according their magnitude. Those, which prove to be quite small, correspond to false exponents, while the others correspond to true ones. With increasing of the number n', the difference between these two groups becomes more distinct. Making formally n' tend to infinity, we come to an idea about replacement of equation (1) by an integral relation of the following form:

$$y(t) = \int_0^{\infty} \alpha(\tau) e^{-t/\tau} d\tau , \tag{5}$$

where τ moves to a continuous variable running all positive real numbers and α moves to a continuous function of this parameter. Then the number of maximums in the distribution $\alpha(\tau)$ and their positions will give us a good estimation of the number of exponential components and the lifetimes. In fact, the obtained integral transformation is nothing but the inverse Laplace transformation. Thus, we reduce our task to a problem of numerical inversion of Laplace transformation.

Generally, to reverse the Laplace transformation, one should solve an integral equation of the first type:

$$\int_0^{\infty} f(x) e^{-px} dx = F(p) , \tag{6}$$

where $F(p)$ is a known function (image) of a complex argument, which proposed to be analytical in a half–plane $\operatorname{Re} p > \gamma$, $\gamma < \infty$. In solving the equation (6) we face with two main problems: necessity to apply a rather sensitive mathematical tools and instability of the original f with respect to image F . In analysing different methods of inversion of Laplace transformation we came to a conclusion that the best variant for us consists in transformation of the initial interval $(0,+\infty)$ of possible values of the argument to a finite one and subsequent orthogonal function system expansion of the original. In practice, we used two

expansions: Jacobi polynomial expansion and Fourier sine–expansion [4]. As above, we assume that we know the function $F(p)$ in points $p = 0,1,\ldots$.
In the first variant we use the function $\omega(y)$, defined as

$$\omega(y) = y^{\alpha}(1-y)^{\beta}, \qquad 0 < y < 1, \qquad \alpha > -1, \qquad \beta > -1, \tag{7}$$

where α and β are assigned before. There exists an orthonormal polynomial system $Q_n(y)$ on the segment $[0,1]$ with the following properties:

$$\int_0^1 Q_n(y)Q_m(y)\omega(y)dy = \delta_{nm} . \tag{8}$$

These polynomials are called "shifted normalised Jacobi polynomials". In the explicit formula for the polynomial $Q_n(y)$

$$Q_n(y) = \sum_{m=0}^{n} \alpha_{nm} y^m \tag{9}$$

the coefficients α_{nm} are well–known (see, for example, [5]). The function $f(x)$ can be represented as

$$f(x) = \phi(e^{-x})\omega(e^{-x})e^{-x} , \tag{10}$$

where

$$\phi(y) = \sum_{n=0}^{\infty} \beta_n Q_n(y) \tag{11}$$

and

$$\beta_n = \sum_{k=0}^{n} F(k) . \tag{12}$$

To realise the second variant we have to know the value $f(0) = f_0$. Then we may reduce our task to the following:

$$\int_0^{\infty} g(x)e^{-px}dx = G(p) , \tag{13}$$

where

$$f(x) = f_0 e^{-x/2} + g(x)e^{-x/2} ,$$

$$G(p+1/2) = F(p) - \frac{f_0}{p+1/2} ,$$

and $g(0) = 0$. An angle θ is defined as $\theta = \arccos(e^{-x/2})$, $0 \le \theta \le \pi/2$, so the function $g(x)$ can be written as $g(x) = \phi(\theta)$, where the function $\phi(\theta)$ is determined by a Fourier series:

$$\phi(\theta) = \sum_{i=0}^{\infty} \alpha_i \sin(2i+1)\theta . \tag{14}$$

The coefficients α_i can be calculated from a triangular linear system:

$$\sum_{k=0}^{n} \frac{2k+1}{2n+1} C_{2n+1}^{n-k} \alpha_k = \frac{2}{\pi} 4^n G(n+1/2) , \qquad n = 0,1,\ldots . \tag{15}$$

Thus, leaving in the expansions (11) or (14) the required number of terms, we can find the original $f(x)$ with a necessary degree of accuracy. In practice, however, we saw that the first algorithm gave us more stable and more precise solution, with appropriate choice of parameters α and β. In conclusion, one can say that all above–listed methods, with small improvements, give us a possibility to distinguish exponential components and to calculate their parameters with enough degree of accuracy, which stimulates further investigations of these algorithms, by means of both analytical and numerical tools.

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