A comparative study of subspace-based methods for 2-D nuclear magnetic resonance spectroscopy signals

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A COMPARATIVE STUDY OF SUBSPACE-BASED METHODS FOR 2-D NUCLEAR MAGNETIC RESONANCE SPECTROSCOPY SIGNALS

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ABSTRACT

One of the major challenges is using subspace-based approaches for the determination of the parameters of one- or multidimensional signals is their practical applicability on real data containing *damped* exponentials. In this paper, we present a comparison survey of low complexity subspace-based methods and we focus on 2-D nuclear magnetic resonance (NMR) spectroscopy application. We first present free search 2-D estimation approaches for damped sinusoids and we analyze their performances using simulated signals. Then, we present the results obtained on real 2-D NMR data.

***Index Terms*—** Two-dimensional NMR, parameter estimation, high-resolution, damped sinusoids.

1. INTRODUCTION

Nuclear magnetic resonance (NMR) spectroscopy is a powerful technique for determining the structure of matter. Multidimensional NMR data can be modeled as a sum of multidimensional damped sinusoids. In the recent decades, numerous high-resolution subspace-based methods have been proposed to estimate the parameters of one-dimensional (1-D) and multidimensional (M-D) signals. For 1-D signals, we can cite the Kumaresan and Tuft approach [1], matrix pencil, ESPRIT, etc. Some method developed for the 2-D case are 2-D TLS-Prony [2], matrix enhancement and matrix pencil (MEMP) [3], 2-D ESPRIT [4], multidimensional folding (MDF) [5], multidimensional embedding (MDE) [6]. Maximum likelihood methods have been shown to achieve the best estimation performance. However they need usually a prohibitive computational complexity to obtain the global maximum of the likelihood function. Among the subspace estimation approaches mentioned, there are methods that apply only to undamped sinusoids. However, the NMR signals consist of damped sinusoids, therefore, they cannot be used directly.

In this paper we present a comparison of free search (low complexity) subspace-based method that can be applied to 2-D NMR data. This comparison is based on the computational complexity and the accuracy of the estimates in terms of their mean square error. Application on real data is also presented. The paper is organized as follows. After a brief description of the signal model of 2-D NMR data, the TLS-Prony, MEMP, 2-D ESPRIT and MDE methods are described in Section 3. Then computer examples are presented in Section 4 and the application of compared methods to experimental NMR data is presented in section 5. Conclusions are drawn in Section 6.

2. 2-D NMR DATA MODEL

NMR data can be described as:

$$y(m,n) = x(m,n) + e(m,n) = \sum_{f=1}^{F} c_f a_f^m b_f^n + e(m,n) \quad (1)$$

for $m = 0,\ldots,M-1$ and $n = 0,\ldots,N-1$, where $a_f = e^{(-\alpha_a + j2\pi f_a)}$ and $b_f = e^{(-\alpha_b + j2\pi f_b)}$, with (α_a, α_b) are damped factors and (f_a, f_b) are frequencies. c_f are complex amplitudes and $e(m,n)$ is a 2-D additive noise. Let $\mathbf{a} = [a_1, a_2, \cdots, a_F]^T$, $\mathbf{b} = [b_1, b_2, \cdots, b_F]^T$, $\mathbf{c} = [c_1, c_2, \cdots, c_F,]^T$. Define two Vandermonde matrices: $\mathbf{A}$ and $\mathbf{B}$ of size $M \times F$ and $N \times F$ with generators $\mathbf{a}$ and $\mathbf{b}$ respectively. We denote $\mathbf{A}_l$ the l first rows of $\mathbf{A}$ and $\mathbf{B}_l$ the l first rows of $\mathbf{B}$.

3. 2-D ESTIMATION ALGORITHMS

3.1. 2-D TLS-Prony Methods

TLS-Prony 2-D estimation method [2] is a generalization of the 1-D TLS Prony. This method decompose the original 2-D estimation problem into two 1-D estimation problems. First, the noiseless data model in (1) can be rewritten as:

$$x(m,n) = \sum_{k=1}^{K} \sum_{l=1}^{L_k} c_{k,l} b_{k,l}^n a_k^m = \sum_{k=1}^{K} h_k(n) a_k^m \quad (2)$$

where $h_k(n) = \sum_{l=1}^{L_k} c_{k,l} b_{k,l}^n$. Therefore, it is obvious that the last term in expression (2) is the 1-D multi-snapshots exponential signals model. Then, the modes $\{a_k\}_{k=1}^{K}$ and the amplitudes $\{h_k(n)\}_{k=1}^K$ for N snapshots are estimated by 1-D TLS-Prony algorithm or other algorithm for estimating 1-D damped exponential signal parameters [7, 8]. Modes and amplitudes $\{b_{k,l}, c_{k,l}\}_{k=1,l=1}^{K,L_k}$ are then estimated applying the same approach to $\{h_k(n)\}$.

In the above algorithm (TLS-Prony1), modes of the second dimension (b-modes) are estimated from estimate of $h_k(n)$. This will cause error propagation and may result in poor b-modes estimates. To avoid this error propagation, a second algorithm (TLS-Prony2) is proposed in which the TLS-Prony1 algorithm is applied first to estimate the a-modes then the b-modes as above, second, it is applied to estimate b-modes, then a-modes. Modes with high accuracy obtained from the two estimations are matched using a distance-based matching algorithm.

The 2-D TLS-Prony algorithms are computationally efficient because TLS-Prony1 resolve twice a 1-D estimation problem and TLS-Prony2 apply twice algorithm1 plus the pairing processing.

3.2. MEMP Method

Matrix Enhancement and Matrix Pencil (MEMP) method was proposed to estimate 2-D modes. To cope rank deficiency of the data matrix, an enhanced matrix Hankel block structure $\mathbf{X}_s$ is formed from the 2-D data:

$$\mathbf{X}_s = \begin{bmatrix} \mathbf{X}_0 & \mathbf{X}_1 & \cdots & \mathbf{X}_{M-K} \\ \vdots & \vdots & & \vdots \\ \mathbf{X}_{K-1} & \mathbf{X}_K & \cdots & \mathbf{X}_{M-1} \end{bmatrix} \tag{3}$$

where

$$\mathbf{X}_m = \begin{bmatrix} x(m,0) & \cdots & x(m,N-L) \\ \vdots & & \vdots \\ x(m,L-1) & \cdots & x(m,N-1) \end{bmatrix} \tag{4}$$

Using (1) in (4), $\mathbf{X}_m$ becames

$$\mathbf{X}_m = \mathbf{B}_L D(\mathbf{c}) D^m(\mathbf{a}) \mathbf{B}_{M-L+1}^T. \tag{5}$$

where $D(\mathbf{c})$ is a square matrix with diagonal elements $\mathbf{c}$. Then, using (5) in (3), $\mathbf{X}_s$ becomes:

$$\mathbf{X}_s = \mathbf{E}_L D(\mathbf{c}) \mathbf{E}_R \tag{6}$$

where

$$\mathbf{E}_L = \begin{bmatrix} \mathbf{B}_L \\ \mathbf{B}_L D(\mathbf{a}) \\ \vdots \\ \mathbf{B}_L D^{K-1}(\mathbf{a}) \end{bmatrix} \tag{7}$$

$$\mathbf{E}_R = [\mathbf{B}_{N-L+1}^T, D(\mathbf{a})\mathbf{B}_{N-L+1}^T, \cdots, D^{M-K}(\mathbf{a})\mathbf{B}_{N-L+1}^T] \tag{8}$$

The singular value decomposition (SVD) of $\mathbf{X}_s$ yields to:

$$\mathbf{X}_s = \mathbf{U}_F \mathbf{S}_F \mathbf{V}_F^H + \mathbf{U}_n \mathbf{S}_n \mathbf{V}_n^H \tag{9}$$

where $\mathbf{U}_F$, $\mathbf{S}_F$ and $\mathbf{V}_F$ contain the F principal components whereas $\mathbf{U}_n$, $\mathbf{S}_n$ and $\mathbf{V}_n$ contain the remaining non-principal components. Estimation of the 2-D modes can then be summarized in three steps:

1. Estimate a-modes from the matrix $D(\mathbf{a})$, i.e., from the eigenvalues of the matrix $\mathbf{F}_1 = \mathbf{U}_1^\dagger \mathbf{U}_2$ where $\mathbf{U}_1 = \mathbf{U}_F$ with the last L rows deleted, and $\mathbf{U}_2 = \mathbf{U}_F$ with the first L rows deleted.
2. Estimate b-modes from the matrix $D(\mathbf{b})$, i.e., from the eigenvalues of the matrix $\mathbf{F}_2 = \mathbf{U}_{1P}^\dagger \mathbf{U}_{2P}$ where $\mathbf{U}_{1P} = \mathbf{P}\mathbf{U}_F$ with the last L rows deleted, and $\mathbf{U}_{2P} = \mathbf{P}\mathbf{U}_F$ with the first L rows deleted. $\mathbf{P}$ is a defined permutation matrix.
3. Pairing the 2-D modes by maximizing the following criterion:

$$J(f,i) = \sum_{t=1}^{F} \| \mathbf{u}_t^H e_L(a_f, b_i) \|^2, \text{ for } f = 1, 2, \ldots, F \tag{10}$$

where $\mathbf{u}_t$ is the tth column of $\mathbf{U}_F$, and $e_L(a_f, b_i) = [1, a_f, \cdots, a_f^{K-1}] \otimes [1, b_i, \cdots, b_i^{L-1}]$.

In this pairing criterion, $e_L(a_f, b_i)$ is a column in $\mathbf{E}_L$ only if the damping factors are null. It can also achieve incorrect pairing when there exist identical modes in certain dimensions. To avoid this problem, another pairing algorithm has been proposed [4]. The MEMP algorithm gives accurate mode estimates and it has the advantage to avoid the polynomial rooting step like in 2-D TLS-Prony algorithms. On the other hand, it requires large memory due to the dimensions of the enhanced matrix.

3.3. 2-D ESPRIT method

The original ESPRIT method is used to estimate the parameters of pure superimposed sinusoids. For damped signals, a 2-D ESPRIT method was proposed in [4] to estimate the frequencies and damping factors. The problem formulation is the same as for MEMP. However, estimation of the 2-D modes is given in the following steps:

1. Eigenvalue decomposition is computed to diagonalize a linear combination of $\mathbf{F}_1$ and $\mathbf{F}_2$:

$$\beta \mathbf{F}_1 + (1-\beta)\mathbf{F}_2 = \mathbf{T}_1^{-1} \mathbf{D} \mathbf{T}_1. \tag{11}$$

2. $\mathbf{D}_a$ and $\mathbf{D}_b$ are obtained by applying transformation $\mathbf{T}_1$ to $\mathbf{F}_1$ and $\mathbf{F}_2$:

$$\begin{cases} \mathbf{D}_a = \mathbf{T}_1 \mathbf{F}_1 \mathbf{T}_1^{-1} \\ \mathbf{D}_b = \mathbf{T}_1 \mathbf{F}_2 \mathbf{T}_1^{-1} \end{cases} \tag{12}$$

3. a-modes and b-modes are extracted from main diagonal of $\mathbf{D}_a$ and $\mathbf{D}_b$, respectively.

It must be noted that the mode vectors **a** and **b** are correctly ordered. This means that the 2-D ESPRIT method does not need a pairing step.

3.4. Multidimensional embedding method

Multidimensional embedding algorithm is an SVD-based 2-D damped sinusoid estimation algorithm. It uses the enhanced data matrix $\mathbf{X}_s$ and data folding, i.e., the 2-D NMR data are folded in a vector

$$\mathbf{x} = [x(0,0), x(0,1), \cdots, x(0,N-1), \\ x(1,0), \cdots, x(M-1,N_1)]^T \qquad (13)$$

Then it can be verified that:

$$\mathbf{x} = (\mathbf{A} \odot \mathbf{B})\mathbf{c}. \qquad (14)$$

The matrices $\mathbf{E}_L$ and $\mathbf{E}_R$ discussed in the MEMP method becomes here:

$$\mathbf{E}_L = \left(\mathbf{A}^{(K)} \odot \mathbf{B}^{(L)}\right) \text{ and } \mathbf{E}_R = \left(\mathbf{A}^{(M-K+1)} \odot \mathbf{B}^{(N-L+1)}\right)^T. \qquad (15)$$

Under the conditions on K and L insuring to the matrix $\mathbf{X}_s$ to be almost rank F, and using the singular values decomposition of the staking of $\mathbf{E}_1$ and $\mathbf{E}_2$ ($\mathbf{E}_1 = \mathbf{E}_L$ with the last rows deleted, $\mathbf{E}_2 = \mathbf{E}_L$ with the first rows deleted), the 2-D modes can be estimated from the eigenvectors instead of the eigenvalues (see [6]). This algorithm achieve automatic mode pairing, but it fails in the case of identical modes in a certain dimension because eigenvectors are not linearly independent anymore.

4. SIMULATION RESULTS

In this section, we present Monte Carlo (300 runs) simulation results to compare performance measured by the mean square error (MSE). The 2-D ESPRIT, MEMP with the new pairing process [4], MDE with only the algebraic step and the two 2-D TLS-Prony algorithms are applied to estimate modes from simulated signals expressed in (1) with 20×20 samples in additive complex Gaussian white noise with variance σ^2. The amplitudes $\{c_f\}_{f=1}^F$ are set to $(3.184, 2.846, 2.846)$. In the first example, we choose three non identical and well separated modes:

$$(f_{1,1}, \alpha_{1,1}; f_{1,2}, \alpha_{1,2}) = (0.10, 0.080; 0.20, 0.075),$$
$$(f_{2,1}, \alpha_{2,1}; f_{2,2}, \alpha_{2,2}) = (0.30, 0.075; 0.10, 0.050),$$
$$(f_{3,1}, \alpha_{3,1}; f_{3,2}, \alpha_{3,2}) = (0.40, 0.050; 0.25, 0.090).$$

Figure 1 shows the MSE of the MDE, 2-D ESPRIT ($\beta = 8$), MEMP, TLS-Prony1–2 algorithms on the estimation of $f_{2,2}$. We plot also associated Cramr-Rao lower bounds (CRB). It can be seen that 2-D ESPRIT and MEMP algorithms exhibit almost the same MSE which is close to the CRB. They have also a better resolution threshold than TLS-Prony algorithms.

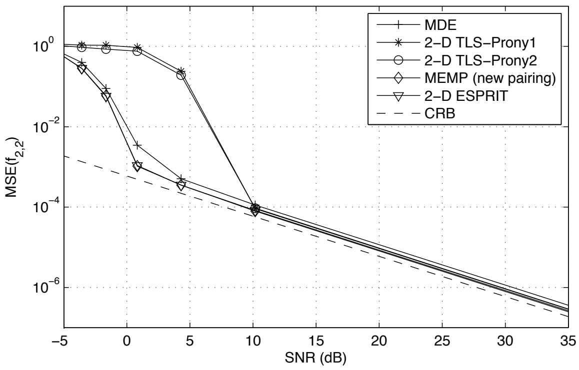

Fig. 1. Performance comparison of the MDE, 2-D ESPRIT, MEMP, TLS-Prony1–2 algorithms. Distinct modes in each dimension.

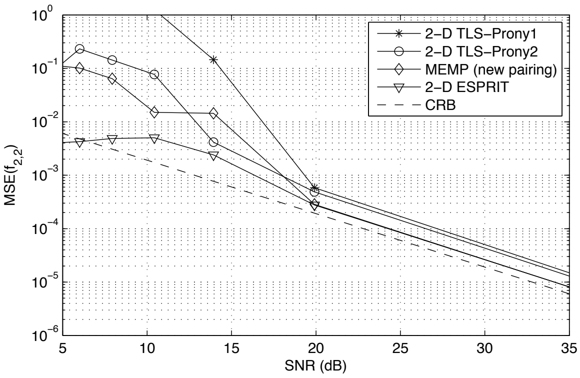

Fig. 2. Performance comparison of the 2-D ESPRIT, MEMP, TLS-Prony1–2 algorithms. Identical and close modes in each dimension.

In the second example, we choose close modes in such a way they cannot be separated by 2-D FFT. Since the resolution limit with 2-D FFT for 20×20 data set is 0.05, simulated modes are:

$$(f_{1,1}, \alpha_{1,1}; f_{1,2}, \alpha_{1,2}) = (0.20, 0.075; 0.20, 0.075),$$
$$(f_{2,1}, \alpha_{2,1}; f_{2,2}, \alpha_{2,2}) = (0.20, 0.075; 0.22, 0.050),$$
$$(f_{3,1}, \alpha_{3,1}; f_{3,2}, \alpha_{3,2}) = (0.22, 0.050; 0.20, 0.075).$$

Note that there exist identical modes along the two dimensions. Therefore, MDE is not applicable in this case. As we can see (figure 2), the results are similar to those of the first example, with the exception that the four algorithms have the same resolution threshold and it is at an SNR of about 20 dB rather than 10 dB for 2-D TLS algorithms and 0 dB for both 2-D ESPRIT and MEMP. These results show that 2-D ESPRIT, MEMP and TLS-Prony1–2 can estimate close and identical modes. Therefore they are good candidates for estimating NMR parameters. However, 2-D ESPRIT and MEMP achieve

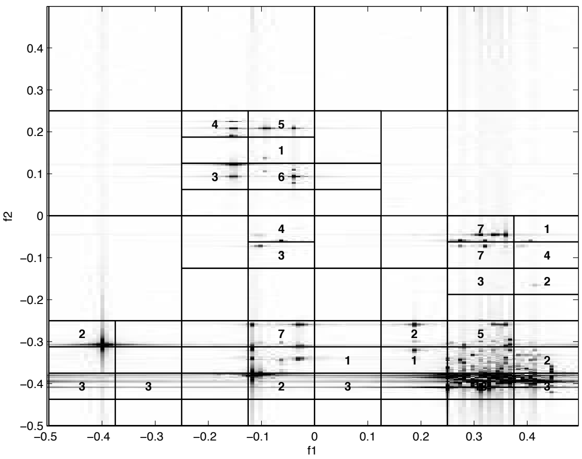

Fig. 3. Magnitude spectrum of the NMR signal. The numbers indicate the number of peaks detected by 2-D ESPRIT.

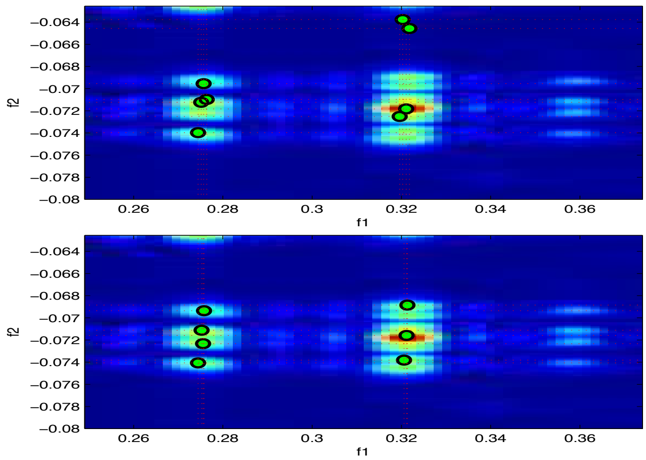

Fig. 4. Results obtained by MDE (top) and 2-D ESPRIT (bottom) in the subband $[0.25, 0.37] \times [-0.10, -0.06]$.

more accurate estimate than TLS-Prony1–2 algorithms.

5. RESULTS ON EXPERIMENTAL DATA

The 2-D ESPRIT and MDE are now compared using an experimental NMR signal. The later correspond to an HMBC experiment (1H–13C correlation by J-coupling). The measured 2048×128 2-D NMR data was obtained obtained from NMR Methodology Laboratory (Nancy, France). Due to the large dimension of the matrix, we used a subband decomposition scheme in order to reduce the computational complexity. The spectrum of the 2-D signal together with the subbands processed are shown on figure 3.

The results obtained by MDE and 2-D ESPRIT are quite similar except the case where exist identical frequencies in a given dimension. For instance, the results obtained by the two methods in the band $[0.25, 0.37] \times [-0.10, -0.06]$ are shown on figure 4. We observe that 2-D ESPRIT succeeds in the detection of all the peaks whereas MDE exhibits an erratic behavior.

6. CONCLUSIONS

In this paper, a comparison survey of two-dimensional low cost estimation method for 2-D NMR spectroscopy signals is presented. Performances of different techniques have been tested using simulation examples. It appeared that 2-D ESPRIT is better than other estimation techniques in terms of the MSE. MDE fails in the case of identical modes in a certain dimension. This is the major limitation for the use of this method for NMR data processing.

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