2DNMR data inversion using locally adapted multi-penalty regularization

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Abstract
Geologists and Reservoir Engineers routinely use time-domain nuclear magnetic resonance (NMR) to learn about the porous structure of rocks that hold underground fluids. In particular, two-dimensional NMR (2DNMR) technique is now gaining importance in a wide variety of applications. Crucial issue in 2DNMR analysis are the speed, robustness and accuracy of the data inversion process. This paper proposes a multi-penalty method with locally adapted regularization parameters for fast and accurate inversion of 2DNMR data. The method solves an unconstrained optimization problem whose objective function contains a data-fitting term, a single \( L_1 \) penalty parameter and a multiple parameter \( L_2 \) penalty. We propose an adaptation of the Fast Iterative Shrinkage and Thresholding (FISTA) method to solve the multi-penalty minimization problem, and an automatic procedure to compute all the penalty parameters. This procedure generalizes the Uniform Penalty principle introduced in [Bortolotti et al., Inverse Problems, 33(1), 2016]. The proposed approach allows us to obtain accurate 2D relaxation time distributions while keeping short the computation time. Results of numerical experiments on synthetic and real data prove that the proposed method is efficient and effective in reconstructing the peaks and the flat regions that usually characterize 2DNMR relaxation time distributions.

Keywords NMR relaxometry · Multi-parameter regularization · Multi-penalty regularization · UPEN2D · FISTA

1 Introduction
Geologists and Reservoir Engineers routinely use time-domain nuclear magnetic resonance (NMR) to learn about the porous structure of rocks that hold underground fluids. NMR is a unique technique that directly senses the presence of \(^1\)H nuclei. Therefore, it permits to accurately estimate many petrophysical parameters, such as porosity, saturation and permeability, etc. Thus nowadays, for example, borehole \(^1\)H NMR is extensively used in oil and gas reservoir characterisation, and recent technological advances have led to tools suitable for environmental applications (see details in [1]). Moreover, two-dimensional NMR (2DNMR) techniques are gaining increasing importance in analysing different porous media ranging from cement to biological tissues. Therefore, an appropriate inversion algorithm of NMR relaxation data is a crucial requirement [4].

In 2DNMR, for example, the joint measurements of the spin relaxation with respect to the longitudinal and transverse relaxation parameters \( T_1 \) and \( T_2 \), respectively, allow us to build two-dimensional relaxation time distributions (\( T_1 - T_2 \) maps). Peaks usually characterize such distributions over flat regions; the position and volume of the peaks are used to obtain information such as petrophysical properties, molecular diffusion, etc [4]. Due to the large dimension of the data and the inherent ill-posedness of the inverse problem, a significant issue in 2DNMR inversion is to ensure both computational efficiency and accuracy. This aspect is particularly relevant in multidimensional logging where
3D NMR inversion algorithms are usually based on methods for 2D NMR inversion [2]. Therefore, the development and application of 3D NMR techniques is seriously restricted by the efficiency and accuracy of the 2D inversion [5]. The measured NMR data $S$ are related to the NMR parameter distribution $F$ according to a Fredholm integral equation of the first kind with separable kernel. For example, in case of $T_1 - T_2$ 2D distributions (2D NMR maps) we have the following continuous model:

$$S(t_1, t_2) = \int_0^\infty k_1(t_1, T_1)k_2(t_2, T_2)F(T_1, T_2)\,dT_1\,dT_2 + e(t_1, t_2)$$

(1)

where $t_1$, $t_2$ are the evolution time parameters and $e(t_1, t_2)$ represents additive Gaussian noise. The kernels $k_1(t_1, T_1)$ and $k_2(t_2, T_2)$ are decaying exponential functions. The discretization of the integral (1) by a quadrature rule leads to a linear inverse problem

$$\textbf{K}f + e = s$$

(2)

where $\textbf{K} = \textbf{K}_2 \otimes \textbf{K}_1$ is the Kronecker product of the discretized decaying exponential kernels $\textbf{K}_1 \in \mathbb{R}^{M_1 \times N_1}$ and $\textbf{K}_2 \in \mathbb{R}^{M_2 \times N_2}$. The vector $s \in \mathbb{R}^N$, $M = M_1 \cdot M_2$, represents the measured noisy signal, $f \in \mathbb{R}^M$, $N = N_1 \cdot N_2$, is the vector reordering of the 2D distribution to be computed and $e \in \mathbb{R}^M$ represents the additive Gaussian noise. The severe ill-conditioning of $\textbf{K}$ is well-known, and it causes the least-squares solution of Eq. 2 to be extremely sensitive to the noise; for this reason, regularization is usually applied. The most common numerical strategies are based on L2 regularization and often use constraints, such as non-negativity constraints, in order to prevent unwanted unphysical results in the computed distribution. This approach requires solving the nonnegatively constrained Tikhonov-like problem:

$$\min_{f \succeq 0} \left\{ \| \textbf{K}f - s \|^2 + \lambda \| f \|^2 \right\}$$

(3)

where $\lambda > 0$ is the regularization parameter. Here and henceforth $\| \cdot \|$ denotes the Euclidean norm. In this context, the approach of Venkataramanan et al. [6], uses data compression to reduce the size of problem (3) and the Butler–Reeds–Dawson method [7] to solve the smaller-size optimization problem. Chouzenoux et al. [8] apply the interior point method for the solution of Eq. 3. The main drawback of single parameter L2 regularization is its tendency to either over-smooth the solution, making it difficult to detect low-intensity peaks, or to under-smooth the solution creating non-physical sharp peaks. Substantial improvements are obtained by the application of multiple parameters Tikhonov regularization, as in the UPEN2D algorithm [10, 11] which solves the minimization problem

$$\min_{f \succeq 0} \left\{ \| \textbf{K}f - s \|^2 + \sum_{i=1}^N \lambda_i (\textbf{L}f)_i^2 \right\}$$

(4)

where $\textbf{L} \in \mathbb{R}^N$ is the discrete Laplacian operator. The multiple regularization parameters $\lambda_i$s are locally adapted, i.e., at each iteration, approximated values for the $\lambda_i$s are computed by imposing the Uniform Penalty (UPEN) principle [10] and a constrained subproblem is solved by the Newton Projection method [12]. Although UPEN2D can obtain very accurate distributions, as reported in the literature [10, 11, 13, 14], its computational cost may be high since it requires the solution of several nonnegatively constrained least-squares problems. In the NMR literature, L1 regularization has been recently considered in order to better reproduce the characteristic sparsity of the relaxation distribution. In [15], the L1 regularization problem

$$\min_{f \succeq 0} \left\{ \| \textbf{K}f - s \|^2 + \alpha \| f \|_1 \right\}$$

(5)

is considered and the Fast Iterative Shrinkage-Thresholding Algorithm (FISTA) [16] is used for its solution. An update searching method is proposed to iteratively determine the regularization parameter as $\alpha = \sqrt{N} \sigma / \| f \|_1$, where $\sigma$ is the standard deviation of the noise. We remark that FISTA is known to be one of the most effective and efficient methods for solving L1-based image denoising and deblurring problems. Recently, FISTA has also been applied to non-convex regularization [17, 18].

The L1 regularization has also been used in NMR to decrease the data acquisition time [19]. In [20], an algorithm related to FISTA is applied to NMR relaxation estimation and comparisons with the methods of Venkataramanan et al. [6], and Chouzenoux et al. [8], are carried out showing the efficiency of the FISTA-like method. However, despite its computational efficiency and its capability of revealing isolated narrow peaks, L1 regularization tends to divide a wide peak or tail into separate undesired peaks.

Recently, the elastic net method [21] with a non-negative constraint

$$\min_{f \succeq 0} \left\{ \| \textbf{K}f - s \|^2 + \lambda \| f \|^2 + \alpha \| f \|_1 \right\}$$

(6)

has been used in [22] to obtain $T_2$ distributions. The problem is formulated as a linearly constrained convex optimization problem and the primal-dual interior method for convex objectives has been applied to one dimensional [22] and two dimensional [23] NMR relaxation problems. However, the performance of the method depends on the two regularization parameters which needs an accurate
tuning; a parameter selection analysis is performed in [23] for a specific set of 2DNMR data.

Our previous review of the current literature shows that for each inversion method, we have to take into account both the efficiency and the accuracy. The UPEN2D method has a great inversion accuracy due to the employment of multi-penalty regularization with locally adapted parameters, but the nonnegative constraints are responsible for its poor computational efficiency. The L1 regularization with FISTA algorithm is computationally very efficient, but its accuracy can be low in the presence of non-isolated peaks. Multi-penalty regularization (6) is able to simultaneously promote distinct features of the sought-for distribution, since it yields a good trade-off among data fitting error, sparsity and smoothness. To overcome the aforementioned drawbacks ensuring both efficiency and accuracy, in this paper, we propose a multi-penalty approach involving L1 and L2 penalties with locally adapted regularization parameters. The proposed method can be mathematically formulated as the unconstrained minimization problem

$$\min_{f} \left\{ \| Kf - s \|^2 + \sum_{i=1}^{N} \lambda_i (L_f)^2 + \alpha \| f \|_1 \right\} \ . \quad (7)$$

This approach allows us to accurately reconstruct distributions with isolated and non-isolated peaks as well as flat areas, in short computation time. On the one hand, L1 regularization prevents from over-smoothing while, on the other hand, local L2 regularization prevents from under-smoothing merging peaks or peak tails. Since L1 regularization enforces sparse distributions, the nonnegative constraints are not included in problem (7) and FISTA can be used for its efficient and effective solution. The UPEN principle is extended to the multi-penalty problem (7) obtaining a very efficient computation strategy for all the regularization parameters. Therefore, tedious multiple parameters tuning procedure is not necessary.

The contribution of this paper is two-fold. Firstly, it introduces a locally adapted multi-penalty model for 2D NMR data inversion, and it proposes an efficient strategy for the automatic computation of the multiple parameters. Secondly, we prove that the solution of Eq. 7 is a regularized solution of problem (2). The extension of the regularization properties of the UPEN principle [Bortolotti et al., Inverse Problems, 33(1), 2016] to a multiple regularization context makes it possible to apply it to more general, non-differentiable and possibly non-convex penalties. The proposed algorithm has been tested on both synthetic and real NMR relaxometry problems, and has been compared to multiple parameters L2 regularization Eq. 4 (UPEN2D) and to L1 regularization (5). The numerical results show the efficiency and effectiveness of the method.

The remainder of the paper is organized as follows: section 2 analyzes the regularization properties of the proposed method. Section 3 reports the details of the numerical algorithm. Finally, in section 4, some results are shown and discussed both on synthetic and real NMR data. The algorithms are implemented in Matlab R2019b and are freely distributed upon request to the authors.

## 2 The uniform penalty principle

In order to generalize to multi-penalty regularization the UPEN principle introduced in [10] for problem (4), let us write problem (7) as

$$\min_{f} \left\{ \| Kf - s \|^2 + \sum_{i=1}^{N+1} \eta_i \phi_i(f) \right\} \quad (8)$$

where

$$\phi_i(f) = \left\{ \begin{array}{ll} (Lf)^2, & i = 1, \ldots, N, \\ \| f \|_1, & i = N + 1, \end{array} \right. \quad \text{and} \quad \eta_i = \left\{ \begin{array}{ll} \lambda_i, & i = 1, \ldots, N, \\ \alpha, & i = N + 1. \end{array} \right. \quad (9)$$

The generalization of the UPEN principle can be stated as follows.

**Definition 2.1 (Generalized Uniform Penalty Principle)** Choose the regularization parameters $\eta_i$ of multi-penalty regularization (8) such that, at a solution $f$, the terms $\eta_i \phi_i(f)$ are constant for all $i$ with $\phi_i(f) \neq 0$, i.e:

$$\eta_i \phi_i(f) = c, \quad \forall i = 1, \ldots, N + 1 \quad \text{s.t.} \quad \phi_i(f) \neq 0 \quad (10)$$

where $c$ is a positive constant.

Let us assume that a suitable bound $\varepsilon$ on the fidelity term of the exact solution $f^\ast$ is given; i.e:

$$\| Kf^\ast - s \|^2 \leq \varepsilon^2 \quad (11)$$

where $f^\ast$ is the solution of the noise-free least-squares problem

$$\min_{f} \left\{ \| Kf - \hat{s} \|^2, \ s = \hat{s} + e \right\}. \quad (12)$$

Following Miller’s criterium [24], the constant $c$ is selected to balance the fidelity and regularization terms in Eq. 8; i.e:

$$c = \frac{\varepsilon^2}{N_0} \quad (13)$$

where $N_0$ is the number of non null terms $\phi_i(f)$:

$$N_0 = \#\{ i \mid \phi_i(f) \neq 0, \ i = 1, \ldots, N + 1 \}. \quad (14)$$

Obviously, with this choice for $c$, Lemma 3.1 of [10] still applies. The lemma is restated here for the sake of clarity.
Lemma 2.1 If $f$ satisfies $\|Kf-s\|^2 \leq \varepsilon^2$ and the parameters $\eta_i$, $i = 1, \ldots, N+1$, are chosen according to the generalized uniform penalty principle with

$$0 \leq c = \frac{\varepsilon^2}{N_0}$$  \hspace{1cm} (15)

where $N_0$ is the number of non null terms $\phi_i(f)$, then

$$0 \leq \|Kf-s\|^2 + \sum_{i=1}^{N_0} \eta_i \phi_i(f) \leq \varepsilon^2 + \sum_{i=1}^{N_0} \frac{\varepsilon^2}{N_0} = 2\varepsilon^2.$$  \hspace{1cm} (16)

Conversely, if $f$ satisfies (16) and the generalized UPEN principle with Eq. 15, then it also satisfies $\|Kf-s\|^2 \leq \varepsilon^2$.

Proof Let $f$ be such that $\|Kf-s\|^2 \leq \varepsilon^2$, then, if Eq. 10 holds with $c$ selected as in Eq. 15, we have

$$0 \leq \|Kf-s\|^2 + \sum_{i=1}^{N_0} \eta_i \phi_i(f) \leq \varepsilon^2 + \sum_{i=1}^{N_0} \frac{\varepsilon^2}{N_0} = 2\varepsilon^2.$$  \hspace{1cm} (17)

Conversely, if Eqs. 16 and 15 hold, then

$$2\varepsilon^2 \geq \|Kf-s\|^2 + \sum_{i=1}^{N_0} \eta_i \phi_i(f) = \|Kf-s\|^2 + \sum_{i=1}^{N_0} \frac{\varepsilon^2}{N_0} = \|Kf-s\|^2 + 2\varepsilon^2.$$  \hspace{1cm} (18)

Hence

$$\|Kf-s\|^2 + \varepsilon^2 \leq 2\varepsilon^2$$

and, by subtraction, we obtain the thesis:

$$\|Kf-s\|^2 \leq \varepsilon^2$$

From Eqs. 10 and 13 we obtain the following expression for the $\eta_i$'s:

$$\eta_i = \frac{\varepsilon^2}{N_0 \phi_i(f)} \text{ for all } i = 1, \ldots, N+1 \text{ such that } \phi_i(f) \neq 0$$

(19)

which can be written in terms of the parameters $\lambda_i$ and $\alpha$ as

$$\lambda_i = \frac{\varepsilon^2}{N_0 (Lf)^T_i} \text{ if } (Lf)_i \neq 0 \text{ and } \alpha = \frac{\varepsilon^2}{N_0 \|f\|_1}.$$  \hspace{1cm} (20)

If the regularization parameters are computed as in Eq. 20, the following lemma shows that the solution of Eq. 7 is a regularized solution of Eq. 2.

Lemma 2.2 Let $f^*$ be the solution of the noise-free least-squares problem (12) and let $f_\varepsilon$ denote the solution to problem (7) where the regularization parameters are chosen according to the generalized uniform penalty principle as follows:

$$\lambda_i = \begin{cases} \frac{\varepsilon^2}{N_0 (Lf)^T_i} \text{ if } (Lf)_i \neq 0; & \text{and } \alpha = \frac{\varepsilon^2}{N_0 \|f\|_1} \\
\gamma \varepsilon^2, & \text{otherwise;}
\end{cases}$$  \hspace{1cm} (21)

where $\gamma$ is a positive constant and $N_0$ is the number of non null terms $(Lf)_i$. Then

$$\lim_{\varepsilon \to 0} f_\varepsilon = f^*$$

and hence $f_\varepsilon$ is a regularized solution of Eq. 2.

Proof Let us define the diagonal matrix $\Lambda$ whose diagonal elements are the parameters $\lambda_i$. The first-order optimality conditions of Eq. 7 are

$$\{0\} \in 2K^T (Kf-s) + 2L^T \Lambda f + \alpha g$$

(22)

where $g$ is the subgradient of $\|f\|_1$, i.e. [3, chp. 3]:

$$g_i = \begin{cases} +1, & \text{if } f_i > 0 \\
-1, & \text{if } f_i < 0, \text{ i.e. } i = 1, \ldots, N. \\
[-1, 1], & \text{if } f_i = 0
\end{cases}$$

In the limit for $\varepsilon \to 0$, from Eq. 20, equation 22 becomes

$$\{0\} \in 2K^T (Kf-s)$$

(23)

which are the first-order optimality conditions for Eq. 12.

\[ \square \]

3 The proposed method

The computation of the parameters $\lambda_i$, $i = 1, \ldots, N$, and $\alpha$ as in Eq. 21 uses the quantities $f^*$ and $\varepsilon$ which are unknown. For this reason, we propose a splitting iterative procedure where they are respectively approximated by the $k$-th iterate $f(k)$ and the corresponding residual norm $\|Kf(k) - s\|$. The proposed iterative procedure is outlined in Algorithm 1 where $\tau$ is the tolerance of the stopping criterium. The parameter $\rho$ is a compliance floor that prevents possible divisions by zero, occurring when $(Lf(k))_i \sim 0$. If $\rho$ is too small the correspondent value $\lambda_i$ may become too large, making the problem very badly conditioned and hence numerically unstable. On the contrary, if $\rho$ is too large, only L1 penalty affects the solution because $\lambda_i \approx 0$.

The computation of each new approximate solution $f(k+1)$ at step 7 of Algorithm 1 is obtained by FISTA [16], after suitable reformulation of the minimization problem. Let us assume that the values $\lambda^{(k)}_i$, $i = 1, \ldots, N$, and $\alpha^{(k)}$ are fixed, then problem (7) can be written as:

$$\min_{f} \{ \Psi_1(f) + \Psi_2(f) \}$$

(24)
Algorithm 1

1: Compute a starting guess $\mathbf{f}^{(0)}$;
2: Choose $\rho, \tau \in (0, 1)$; set $k = 0$;
3: repeat
4: Set $\epsilon^{(k)} = \|\mathbf{Kf}^{(k)} - \mathbf{s}\|^2$
5: Set $\lambda_i^{(k)} = \frac{1}{(N+1)\epsilon^{(k)}}$, $i = 1, \ldots, N$
6: Set $\alpha^{(k)} = \frac{\epsilon^{(k)}}{N+1}\|\mathbf{f}^{(k)}\|_1$
7: Compute $\mathbf{f}^{(k+1)} = \arg\min_{\mathbf{f}} \left\{ \|\mathbf{Kf} - \mathbf{s}\|^2 + \sum_{i=1}^{N} \lambda_i^{(k)} (\mathbf{Lf})_i^2 + \alpha^{(k)} \|\mathbf{f}\|_1 \right\}$
8: Set $k = k + 1$
9: until $\|\mathbf{f}^{(k+1)} - \mathbf{f}^{(k)}\| \leq \tau \|\mathbf{f}^{(k)}\|

where:

$$\psi_1(\mathbf{f}) = \left( \begin{bmatrix} I & 0 \\ 0 & \sqrt{\lambda} \mathbf{I} \end{bmatrix} \right)^{\frac{1}{2}} (\mathbf{K} \mathbf{f} - \begin{bmatrix} \mathbf{s} \\ 0 \end{bmatrix})^2,$$

$I \in \mathbb{R}^{M \times M}$ is the Identity matrix, $\mathbf{0} \in \mathbb{R}^{M \times N}$ is a zero block, and

$$\psi_2(\mathbf{f}) = \alpha^{(k)} \|\mathbf{f}\|_1.$$

The FISTA steps for the solution of Eq. 24 are reported in Algorithm 2 where $\xi$ is a constant stepsize and the starting guess corresponds to solution computed in Algorithm 1 at the $k$-th step.

Algorithm 2 – $\mathbf{f}^{k+1} = \text{FISTA\_STEP}(\xi, \mathbf{f}^{k}, \psi_1, \psi_2)$.

1: Set $t_0 = 1$; $j = 0$; $\mathbf{y}^{(1)} = \mathbf{f}^{k}$
2: repeat
3: $j = j + 1$
4: $\mathbf{f}^{(j)} = \arg\min_{\mathbf{f}} \left\{ \psi_2(\mathbf{f}) + \frac{\xi}{2} \|\mathbf{f} - \mathbf{y}^{(j)} - \frac{1}{\xi} \nabla(\psi_1(\mathbf{y}^{(j)}))\|^2_2 \right\}$
5: $t_{j+1} = \frac{1}{2} \left( 1 + \sqrt{1 + 4t_j^2} \right)$
6: $\mathbf{y}^{(j+1)} = \mathbf{f}^{(j)} + \frac{(t_{j+1} - 1)}{t_{j+1}} (\mathbf{f}^{(j)} - \mathbf{f}^{(j-1)})$
7: until stopping criterion
8: $\mathbf{f}^{(k+1)} = \mathbf{f}^{(j+1)}$

The FISTA iterations are stopped when the relative distance between two successive values of the objective function of Eq. 24 becomes smaller than a given tolerance value $\epsilon^{\text{FISTA}}$. At step 4 of Algorithm 2, the components of $\mathbf{f}^{(j)}$ are computed explicitly, element-wise, by means of the soft thresholding operator:

$$\mathbf{f}_i^{(j)} = \text{sign} \left( z_i^{(j)} - \frac{\alpha}{\xi} \right) \max \left( |z_i^{(j)}| - \frac{\alpha}{\xi}, 0 \right), \quad i = 1, \ldots, N$$

where

$$z_i^{(j)} = \mathbf{y}_i^{(j)} - \frac{1}{\xi} \nabla(\psi_1(\mathbf{y}^{(j)})).$$

The convergence of FISTA has been proven for any stepsize $\xi$ such that $\xi \geq \mathcal{L}(\psi_1)$, where $\mathcal{L}(\psi_1)$ is the Lipschitz constant for the gradient $\nabla \psi_1$ [16]; i.e:

$$\mathcal{L}(\psi_1) = \lambda_{\max}(\mathbf{K}^T \mathbf{K} + \mathbf{L}^T \mathbf{L})$$

where $\lambda_{\max}(\mathbf{X})$ represents the maximum eigenvalue of the matrix $\mathbf{X}$.

The following theorem shows that an upper bound for $\mathcal{L}(\psi_1)$ can be easily provided, thus obtaining the convergence of FISTA.

**Theorem 3.1** Let $\sigma_1^{(1)}$ and $\sigma_1^{(2)}$ be the maximum singular values of the matrices $\mathbf{K}_1$ and $\mathbf{K}_2$, respectively, and let $\lambda_i^{(k)}$ be the local regularization parameters computed at $kth$ step of Algorithm 1, then the value $\xi$ defined as follows:

$$\xi = (\sigma_1^{(1)} \sigma_1^{(2)})^2 + 64 \max_i \lambda_i^{(k)}$$

satisfies

$$\xi \geq \mathcal{L}(\psi_1)$$

and it guarantees the convergence of the FISTA method.

**Proof** We recall that the spectral norm of the symmetric matrix $\mathbf{K}^T \mathbf{K} + \mathbf{L}^T \mathbf{L}$ is defined as

$$\|\mathbf{K}^T \mathbf{K} + \mathbf{L}^T \mathbf{L}\|_2 = \lambda_{\max}(\mathbf{K}^T \mathbf{K} + \mathbf{L}^T \mathbf{L}).$$

Then, by using the triangular inequality,

$$\|\mathbf{K}^T \mathbf{K} + \mathbf{L}^T \mathbf{L}\|_2 \leq \|\mathbf{K}^T \mathbf{K}\|_2 + \|\mathbf{L}^T \mathbf{L}\|_2$$

we obtain

$$\lambda_{\max}(\mathbf{K}^T \mathbf{K} + \mathbf{L}^T \mathbf{L}) \leq \lambda_{\max}(\mathbf{K}^T \mathbf{K}) + \lambda_{\max}(\mathbf{L}^T \mathbf{L})$$

since $\mathbf{K}^T \mathbf{K}$ and $\mathbf{L}^T \mathbf{L}$ are symmetric matrices.

Using the Kronecker product properties of the Singular Value Decomposition (SVD), we have:

$$\lambda_{\max}(\mathbf{K}^T \mathbf{K}) = \lambda_{\max}((\mathbf{K}_2 \otimes \mathbf{K}_1)^T (\mathbf{K}_2 \otimes \mathbf{K}_1)) = (\sigma_1^{(1)} \sigma_1^{(2)})^2.$$ 

(27)

Concerning the term $\lambda_{\max}(\mathbf{L}^T \mathbf{L})$ we can apply the property of the discrete Laplacian matrix $\lambda_{\max}(\mathbf{L}) \leq 8$. This property easily follows from a structured finite difference representation of the Laplace operator with unit step size. Hence:

$$\lambda_{\max}(\mathbf{L}^T \mathbf{L}) \leq 64 \max_i |\lambda_i^{(k)}|.$$ 

(28)

Finally, collecting the terms (27) and (28), we obtain the value $\xi$ that guarantees the convergence of FISTA steps. 

$\square$
We observe that, in NMR, the matrices \( K_1 \) and \( K_2 \) are usually of small size and their SVD can be easily performed in order to compute the values \( \sigma_1 \) and \( \sigma_2 \).

Following the observations in [10], we apply Algorithm 1 with the following \( L_2 \) penalty parameters, which have proven to be very efficient with NMR problems:

\[
\lambda_{i}^{(k)} = \frac{\|Kf^{(k)} - s\|^2}{(N + 1) \left( \beta_0 + \beta_p \max_{\mu \in I_i} (p_{\mu}^{(k)})^2 + \beta_c \max_{\mu \in I_i} (c_{\mu}^{(k)})^2 \right)}, \quad i = 1, \ldots, N
\]

(29)

where

\[ c^{(k)} = Lf^{(k)}, \quad p^{(k)} = \text{vec} \left( \|\nabla F^{(k)}\| \right), \quad f^{(k)} = \text{vec} (F^{(k)}) \]

and \( F^{(k)} \) is the \( k \)-th distribution map (here, \( \text{vec}(V) \) denotes the vector obtained by columnwise reordering the elements of a matrix \( V \)). The \( I_i \) are the indices subsets related to the neighborhood of the point \( i \) and the \( \beta \)'s are positive parameters; \( \beta_0 \) prevents division by zero and is a compliance floor, which should be small enough to prevent over-smoothing. The optimum value of \( \beta_0, \beta_c \) and \( \beta_p \) can change with the nature of the measured sample.

Finally, the proposed procedure is stated in Algorithm 3 and is called L1LL2 method, which comes from “method based on \( L_1 \) and Locally adapted \( L_2 \) penalties”. As already discussed in [10], the starting guess \( f^{(0)} \) is computed by applying a few iterations of the Gradient Projection method to the nonnegatively constrained least squares problem

\[ \min_{F \geq 0} \|KF - s\|^2. \]

### 4 Numerical results

The analysis of the proposed algorithm is carried out in this section both on synthetic and real NMR relaxation data. The numerical tests are performed on a PC laptop equipped with 2.9 GHz Intel Core i7 quad-core, 16 GB RAM. The algorithms are implemented in Matlab R2019b. The values \( \tau = 10^{-3} \) and \( \tau_{\text{FISTA}} = 10^{-7} \) have been fixed for the stopping tolerances of L1LL2 (Algorithm 3) and FISTA (Algorithm 2) respectively.

#### 4.1 Synthetic data

Aim of this paragraph is to analyse the accuracy and performance of the proposed L1LL2 algorithm. To this purpose we test L1LL2 algorithm on synthetic data that emulates the results of measurement with a 2D IR-CPMG sequence (see [10, 13, 14]), where the kernels of Eq. 1 are denoted as follows:

\[ s = Kf^* + e. \]

The tests are carried out by running 10 noise realizations with \( \delta = 10^{-2} \) and the numerical results reported in the Tables 1 and 2 are averaged over these noise realizations.

\[
\text{Algorithm 3 – L1LL2 method.}
\]

1. Choose \( \tau \in (0, 1) \) and \( \beta_0, \beta_p, \beta_c > 0 \)
2. Set \( k = 0 \) and compute \( f^{(0)} \)
3. Compute \( \sigma_1^{(1)} \) and \( \sigma_1^{(2)} \)
4. \text{repeat}
5. Set \( \varepsilon^{(k)} = \|KF^{(k)} - s\|^2 \)
6. Set
7. \text{compute}
8. Set \( \xi^{(k)} = \left( \sigma_1^{(1)} \sigma_1^{(2)} \right)^2 + 64 \max_i |\lambda_i^{(k)}| \)
9. \text{Compute}
10. \text{Set}
11. \text{until} \|f^{(k+1)} - f^{(k)}\| \leq \tau \|f^{(k)}\|

Two different predefined relaxation maps \( F(T_1, T_2) \) are used to compute two set of synthetic relaxation data.

The first relaxation map, named 2PKs test, has size \( N_1 \times N_2 \) where \( N_1 = N_2 = 80 \). The relaxation map, represented in Fig. 1a, has two peaks at positions \( (T_1 = 814.97 \, ms, T_2 = 4.533 \, ms) \) and \( (T_1 = 119.54 \, ms, T_2 = 8.5606 \, ms) \), relative to two population of spins with the NMr constraint \( T_1 > T_2 \). The second relaxation map, named 3PKs test, has 100 \times 100 points. In this case the relaxation map, represented in Fig. 1b, has three peaks, relative to three population spins, with peaks in the following positions: \( (T_1 = 1582.2 \, ms, T_2 = 32.289 \, ms) \), \( (T_1 = 5.9692 \, ms, T_2 = 2.6124 \, ms) \) and \( (T_1 = 1139.5 \, ms, T_2 = 258.08 \, ms) \). In both cases the number of the IR points is \( M_1 = 128 \), while for CPMG it is \( M_2 = 2048 \).

Normal Gaussian random noise \( e \) of level \( \delta \equiv ||e|| \) is added as follows:

\[ s = Kf^* + e. \]

The tests are carried out by running 10 noise realizations with \( \delta = 10^{-2} \) and the numerical results reported in the Tables 1 and 2 are averaged over these noise realizations.
The algorithms accuracy is measured by means of the relative error $E_{rel}^2$ and the Root Mean Squared Deviation $RMSD$, defined as follows:

$$E_{rel}^2 = \frac{\|f - f^*\|^2}{\|f^*\|^2}, \quad RMSD = \frac{\|\hat{s} - s\|}{\sqrt{M}}, \quad \hat{s} = Kf$$

where $f$ represents the map computed by the algorithms and $f^*$ is the true map. The first analysis evaluates the effects of the multi-penalty and multi-parameter approach (L1LL2) compared to the L1 and multi-parameter L2 penalties. The adapted L1 penalty algorithm (A-L1) is obtained applying the algorithm 1 to solve (7) with $\lambda_i = 0$, while multi-parameter L2 is obtained by solving problem (3) with algorithm UPEN2D. In Table 1, the error parameters and computation times are reported for each algorithm. While UPEN2D reaches always the most accurate solutions, the A-L1 algorithm has the greatest relative error values. Conversely, A-L1 is the fastest method while UPEN2D requires the longest computation times. We observe that L1LL2 achieves the best trade-off between accuracy and computation time. We can quantify such trade-off in terms of Percentage Accuracy Loss (PAL), defined as

$$PAL_m = \frac{Err_m - Err_{min}}{Err_{min}} \times 100$$

where $Err_m$ represents the relative error of method $m$ (L1LL2 or A-L1) and $Err_{min}$ is the minimum relative error, always obtained by UPEN2D. Analogously, we measure the Percentage Efficiency Gain (PEG) by subtracting the computation time of L1LL2 or A-L1 ($Time_m$) to that of UPEN2D ($Time_{max}$):

$$PEG_m = \frac{Time_{max} - Time_m}{Time_{max}} \times 100$$

The values reported in Table 2 show that for the 2pks test the accuracy lost by L1LL2 is about 22% smaller than A-L1 while the computation efficiency gained is similar. Concerning the 3Pks test we observe that accuracy lost by L1LL2 is about 25% smaller than A-L1 while the performance gained by A-L1 is 11% greater than

| Test | Algorithm | $E_{rel}^2$ | $RMSD$ | Time |
|------|-----------|-------------|--------|------|
|      |           | (-)         | (a.u.) | (s)  |
| 2Pks | L1LL2     | 1.22 10^{-1}| 1.953 10^{-4}| 10.80|
|      | A-L1      | 1.41 10^{-1}| 1.953 10^{-4}| 10.13|
|      | UPEN2D    | 8.79 10^{-2}| 1.953 10^{-4}| 68.6
|      | L1LL2     | 1.09 10^{-1}| 1.381 10^{-4}| 29.00|
| 3Pks | A-L1      | 1.31 10^{-1}| 1.381 10^{-4}| 19.84|
|      | UPEN2D    | 8.51 10^{-2}| 1.381 10^{-4}| 85.62|

Reference value $RMSD^* = \delta/\sqrt{M} = 1.9531 10^{-5}$
Table 2 Percentage Accuracy Loss (PAL (32)) and Percentage Efficiency Gain (PEG (33)) obtained by each method on the two different test problems

| Test | Algorithm | PAL (%) | PEG (%) |
|------|-----------|---------|---------|
| 2Pks | L1LL2     | 38.8    | 84.3    |
|      | A-L1      | 60.5    | 85.2    |
| 3Pks | L1LL2     | 28.1    | 66.1    |
|      | A-L1      | 53.9    | 78.0    |

that of L1LL2. Hence L1LL2 reaches the best balance between accuracy and computation time. This feature is well represented in Fig. 2 where the time evolution of $E_{rel}^2$ is plotted for each algorithm. We observe that the introduction of the $L_1$ regularization causes a considerable decrease in the total computation time at the expenses of a slight increase in the relative error.

The contribution of the multi-parameter $L_2$ term to the algorithm accuracy is further highlighted in Figs. 3 and 4 where the maps computed by the three algorithms on the 2Pks and 3Pks tests are shown. We observe that figures (b) and (d) are more precise in reproducing the true contour levels (figure (a)), compared to the A-L1 regularization algorithm in figure (c).

Concerning the residual values, we observe that the RMSD parameters reported in Table 1 have indeed very tiny differences, in the range $[10^{-8}, 10^{-7}]$, revealing equal data consistency for all methods. Moreover the good results are confirmed by the RMSD correspondent to the true solution $f^*$, given by $RMSD^* = \delta/\sqrt{M} = 1.9531 \times 10^{-5}$.

4.2 Real data

In this section we report the reconstructions obtained with L1LL2 on real data acquisitions. Two measured samples are studied: a White Portland Cement (WPC) and a Maastricht stone, a limestone characterized by high porosity [9], (see [11], for a detailed description of acquisition parameters and instrument characteristics).

The first test, named $T_1 - T_2$ test, is relative to an $^1$H IR-CPMG measurement performed on a WPC sample hardened a few tens of minutes. The kernels are defined in Eq. 30, the data set have $48 \times 1000$ elements and the computed relaxation time map has $80 \times 80$ points. The second test, named $T_2 - T_2$ test is related to a $^1$H CPMG-CPMG measurement performed on a Maastricht stone full saturated with water. In this case, both kernels $k_1$ and $k_2$ refer to transversal relaxation times $T_{21}, T_{22}$ with the following expression:

$$k_1(t_1, T_{21}) = \exp(-t_1/T_{21}), \quad k_2(t_2, T_{22}) = \exp(-t_2/T_{22}).$$

and the $T_1, T_2$ in model (1) are substituted by $T_{21}, T_{22}$. The reconstructed relaxation time map has $64 \times 64$ points while the acquired data set has $128 \times 2800$ elements. Since in previous computations UPEN2D yielded the most accurate solutions on data set coming from different sequences and samples (see [10, 13, 14] and [11]), we use it as a reference method and we evaluat the following relative error $E_{relUPEN2D}$:

$$E_{relUPEN2D} = \frac{\|f - f_{UPEN2D}\|}{\|f_{UPEN2D}\|}$$

From the results reported in Table 3 we observe that despite the similarity of the RMSD values within the same
test problem, there is a remarkable difference in the relative distance from the reference solution computed by UPEN2D (column $E_{\text{rel}}^{UPEN2D}$). Indeed $E_{\text{rel}}^{UPEN2D}$ of A-L1 is always greater than that obtained by L1LL2. Moreover, the computation times show the improved efficiency of L1LL2, as expected. In case of the $T_1 - T_2$ test, analysing the computed 2D contour map, Fig. 5, we can see two peaks with $T_2$ values at about 0.8 ms and 20 ms. As it well known, WPC pore structures depend on the sample preparation, and the hydration processes cause their continuously evolution over time, therefore these two peaks seem to be compatible with the population of spins into interhydrate spaces and capillary pores, respectively [25]. A small artifact is visible in the contour map (Fig. 5) as well as in the T1 projection (Fig. 6). As a general comment, we can see the optimal correspondence of L1LL2 with UPEN2D in the 1D maps projections reported in Fig. 6 where both peaks are well defined in position, height and amplitude. The contour maps of the computed 2D relaxation time distributions, reported in Fig. 5, also confirm the good accuracy of L1LL2 compared to UPEN2D. We observe in Fig. 5c the poor A-L1 reconstruction, probably due to the low S/N ratio of the acquired data.

Concerning the $T_2 - T_2$ test, we again observe in Table 3 the preservation of consistency of the results between L1LL2 and UPEN2D algorithms. The relaxation time contour maps, reported in Fig. 7, report a precise reproduction of spin population, confirmed also by the projection on horizontal axes in Fig. 8b. Two-dimensional $T_2 - T_2$ exchange experiments are useful to assess exchange between different relaxation centers and to understand the pore connectivity. Off-diagonal peaks in Fig. 7 indicate
Fig. 4 Contour plots of the 3pks test. a True distribution map, b L1LL2 reconstruction, c A-L1 reconstruction, d UPEN2D reconstruction

Table 3 RMSD, $E_{relUPEN2D}$ and computation times obtained with 2D NMR real data

| Test  | Algorithm | RMSD   | $E_{relUPEN2D}$ | Time |
|-------|-----------|--------|----------------|------|
| $T_1 - T_2$ | L1LL2     | 3.33 $10^{-3}$ | 9.36 $10^{-2}$ | 4.2  |
|       | A-L1      | 3.03 $10^{-3}$ | 3.91 $10^{-1}$ | 6.5  |
|       | UPEN2D    | 3.36 $10^{-3}$ | -              | 54.6 |
| $T_2 - T_2$ | L1LL2     | 1.248 $10^{-1}$ | 1.99 $10^{-1}$ | 4.32 |
|       | A-L1      | 1.235 $10^{-1}$ | 5.00 $10^{-1}$ | 7.85 |
|       | UPEN2D    | 1.251 $10^{-1}$ | -              | 10.6 |
Fig. 5 Test $T_1 - T_2$, contour maps of relaxation times. 

- (a) computed in 54.6 s. 
- (b) computed in 3.53 s. 
- (c) computed in 6.6 s.

Fig. 6 $T_1 - T_2$ test, projection of the time relaxation maps onto the vertical axis $T_1$ and onto the horizontal axis $T_2$. L1LL2 red dashed line, UPEN2D blue line.
Fig. 7 Test $T_2 - T_2$, contour maps of relaxation times. 
\(\text{a}\) computed in 10.3 s, \(\text{b}\) computed in 3.6 s \(\text{c}\) computed in 7.8 s

Fig. 8 T2-T2 test, projection of the time relaxation maps onto the vertical axis $T_{21}$ and onto the horizontal axis $T_{22}$. L1LL2 red dashed line, UPEN2D blue line
5 Conclusion

This paper presents the L1LL2 method for the inversion of 2D NMR relaxation data. The algorithm automatically computes a 2D distribution map of NMR parameters and spatially adapted regularization parameters by iteratively solving a sequence of multi-penalty problems. The FISTA method is used for the solution of the minimization problems, and all the regularization parameters are updated according to the uniform penalty principle. The L1LL2 method has been compared to A-L1 and UPEN2D algorithms. The numerical results show that, compared to UPEN2D, the main advantage of L1LL2 is the increased computational speed without a significant loss in the inversion accuracy; compared to A-L1, L1LL2 provides more accurate distributions at a comparable computational cost. Future work will consider the extension of this method to different penalty functions to employ the generalized uniform Penalty principle to a broader variety of inverse problems.

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