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Abstract. This paper is inspired by recently proposed approach for interpreting data of Electrochemical Impedance Spectroscopy (EIS) in terms of Distribution of Diffusion Times (DDT). Such interpretation requires to solve a Fredholm integral equation of the first kind, which may have a non-square-integrable kernel. We consider a class of equations with above-mentioned peculiarity and propose to regularize them in weighted functional spaces. One more issue associated with DDT-problem is that EIS data are available only for a finite number of frequencies. Therefore, a regularization should unavoidably be combined with a collocation. In this paper we analyze a regularized collocation in weighted spaces and propose a scheme for its numerical implementation. The performance of the proposed scheme is illustrated by numerical experiments with synthetic data mimicking EIS measurements.

1 Introduction

Electrochemical Impedance Spectroscopy (EIS) has become a powerful technique to determine resistive and dielectric properties of materials [1]. In EIS, the angular excitation frequency ω of the applied potential is varied over a range of frequencies $[\omega_1, \omega_m]$, while the impedance (both resistance and inductance) is measured.

The traditional interpretation of EIS data is given in terms of a distribution of relaxation times (DRT) that maps impedance spectra on a function containing the timescale characteristics of the system under consideration [3]. As it is pointed out in [17], DRT analysis is intended only to represent high-frequency region of the impedance spectra (i.e., interfacial charging and Faradaic reactions).

At the same time, low-frequency impedance may contain geometrical information that can be used to infer microstructural statistics of heterogeneous and nanostructural materials. A theory of such impedance for random heterogeneous materials has been recently proposed in [17]. It is based on the concept of a function of distribution of diffusion times (DDT) and is given by Fredholm integral equations

$$y(\omega) = \int_0^\infty K(\omega, \tau) p(\tau) \, d\tau, \tag{1}$$

where $p(\tau)$ is DDT, $y(\omega)$ is the collective diffusion admittance, and $K(\omega, \tau)$ is the so-called finite-length diffusion model that represents individual diffusion paths depending on boundary conditions and symmetries.

When inverting the impedance spectrum $y(\omega)$ to obtain DDT $p(\tau)$, one should be aware that this class of inversion problems is known to be mathematically ill-posed and, thus, should be treated with a regularization.

Moreover, since only a finite number of spectrum measurements $y(\omega_i)$, $i = 1, 2, \ldots, m$, may actually be performed, the above mentioned equations (1) appear as after application of a collocation method that needs to be combined with the regularization.

In EIS literature such a combination has been discussed in the context of inversion of DRT equations [15], [16], [5], [22], [2], [4], [24].

One of the essential differences between DRT and DDT approaches is that, in contrast to DRT equation, the kernels $K(\omega, \tau)$ in (1) may not be square-integrable on $[\omega_1, \omega_m] \times [0, \infty)$. For example, the real and imaginary parts of the kernel $K(\omega, \tau) = \sqrt{i\omega\tau}\tanh(\sqrt{i\omega\tau})$ suggested in [17] for blocking boundary conditions and planar symmetry, asymptotically behave like $O(\sqrt{\omega\tau})$ and clearly are not square-integrable on $[\omega_1, \omega_m] \times [0, \infty)$. Then for the equations (1) with such kernels $K(\omega, \tau)$ a straightforward application of Tikhonov regularization combined with the discretization by the trapezoidal rule, as it is adopted in [17], is not justified by the Regularization theory.

Therefore, the goal of the present study is to theoretically analyze the regularized collocation of DDT-problems. In the next section we consider somewhat more general class of equations (1), containing DDT equations as special cases, and study their regularization in weighted L_2-spaces. Then we interprete our results in the context of DDT-problem and illustrate them by numerical examples with synthetic data.

2 A class of integral equations with non-integrable kernels and their regularization

Let $L_{2,\infty} = L_2(0, \infty)$ be the Hilbert space of functions f that are square-integrable over half line $[0, \infty)$,

$$\|f\|^2_{L_{2,\infty}} = \int_0^\infty |f(\tau)|^2 \, d\tau < \infty.$$

By $L_2^\beta = L_2^\beta(0, \infty)$, $\beta > 0$, we denote a weighted subspace of $L_{2,\infty}$ with the norm

$$\|f\|_{L_2^\beta} := \left(\int_0^\infty (1+\tau)^{2\beta} |f(\tau)|^2 \, d\tau \right)^{1/2} < \infty.$$

In the sequel the integral operators

$$Kp(\omega) = \int_0^\infty K(\omega, \tau) p(\tau) d\tau, \qquad \omega \in [\omega_1, \omega_m],$$

in (1) will be considered as acting from L_2^β into the Hilbert space $L_{2,\Omega} = L_2(\omega_1, \omega_m)$ of functions y that are square-integrable over the interval (ω_1, ω_m),

$$\|y\|_{L_{2,\Omega}}^2 = \int_{\omega_1}^{\omega_m} |y(\omega)|^2 \, d\omega < \infty.$$

We will need also an embedding operator J of L_2^β into $L_{2,\infty}$ such that for any $f \in L_2^\beta$ we have

$$Jf(\tau) = (1+\tau)^\beta f(\tau) \in L_{2,\infty}.$$

The adjoint of J is the operator $J^* : L_{2,\infty} \to L_2^\beta$ assigning to a function $g \in L_{2,\infty}$ the function

$$J^* g(\tau) = (1+\tau)^{-\beta} g(\tau) \in L_2^\beta.$$

Then the equation (1) can be rewritten as

$$Hx = y, \tag{2}$$

where

$$x(\tau) = Jp(\tau) = (1+\tau)^\beta p(\tau) \in L_{2,\infty}$$

and H is an integral operator

$$Hx(\omega) = \int_0^\infty H(\omega,\tau) x(\tau) d\tau, \qquad \omega \in [\omega_1, \omega_m], \tag{3}$$

with a kernel

$$H(\omega,\tau) = K(\omega,\tau)(1+\tau)^{-\beta}. \tag{4}$$

In what follows we assume that β can be chosen in such a way that the kernel $H(\omega,\tau)$ is square-integrable on $[\omega_1, \omega_m] \times [0, \infty)$. For example, as it will be shown in the next section, the kernel $K(\omega,\tau) = \sqrt{i\omega\tau}\tanh(\sqrt{i\omega\tau})$ considered in the paper [17] providing the motivation for the present study, allows the choice $\beta > 3/2$ to make the corresponding kernels (4) square-integrable. Note that the operator (3), (4) with the above chosen β is a compact operator from $L_{2,\infty}$ into $L_{2,\Omega}$. Therefore, potentially one could employ standard regularization techniques to treat the equation (2), but since in the application we have in mind, only a finite number of measurements $y(\omega_j)$, $j = 1, 2, \ldots, m$, is available, the equation (2) is given only in a collocated form.

Formally, it can be written in terms of the sampling operator $S_\Omega g = (g(\omega_1), g(\omega_2), \ldots, g(\omega_m)) \in \mathbb{R}^m$ as

$$S_\Omega H x = S_\Omega y, \tag{5}$$

where the image space $\mathbb{R}^m$ of the operator S_Ω is the Euclidean space of vectors $u = (u_1, \ldots, u_m)$, $v = (v_1, \ldots, v_m)$, equipped with the inner product

$$\langle u, v \rangle_{\mathbb{R}^m} = \sum_{j=1}^{m} \gamma_j u_j v_j, \quad \gamma_j > 0, \quad j = \overline{1, m}.$$

Regularizing (5) by Tikhonov method we arrive at the equation

$$\alpha x + H_\Omega^* H_\Omega x = H_\Omega^* S_\Omega y, \tag{6}$$

where

$$H_\Omega = S_\Omega H = S_\Omega K J^* : L_{2,\infty} \to \mathbb{R}^m,$$

$$H_\Omega x = \left(\int_0^\infty K(\omega_i, \tau)(1+\tau)^{-\beta} x(\tau) \, d\tau \right)_{i=1}^{m}.$$

The solution $x_{\alpha,m}$ of the regularized equation (6) admits the representation

$$x_{\alpha,m} = (\alpha I + H_\Omega^* H_\Omega)^{-1} H_\Omega^* S_\Omega y. \tag{7}$$

Moreover, it is easy to check that

$$H_\Omega^* u(\tau) = \sum_{j=1}^{m} \gamma_j u_j K(\omega_j, \tau)(1+\tau)^{-\beta}.$$

Then the regularized solution has the form

$$x_{\alpha,m}(\tau) = \sum_{j=1}^{m} x_\alpha^j K(\omega_j,\tau)(1+\tau)^{-\beta}, \tag{8}$$

where the coefficients x_α^j solve the following system of linear algebraic equations

$$\alpha x_\alpha^j + \gamma_j \sum_{l=1}^{m} x_\alpha^l \int_0^\infty K(\omega_j,v)K(\omega_l,v)(1+v)^{-2\beta}\, dv = \gamma_j y(\omega_j), \quad j=\overline{1,m}. \tag{9}$$

In practice, however, instead of $S_\Omega y = (y(\omega_1), y(\omega_2), \ldots, y(\omega_m))$ only noisy measurements $(y^\delta(\omega_1), y^\delta(\omega_2), \ldots, y^\delta(\omega_m))$ of $y(\omega)$ are available. Formally, a vector of such measurements can be seen as the image $S_\Omega y^\delta$ of some perturbed version $y^\delta(\omega)$ of data function $y(\omega)$. Then the level of perturbations can be estimated as follows

$$\|S_\Omega(y-y^\delta)\|^2_{\mathbb{R}^m} := \sum_{j=1}^{m} \gamma_j |y(\omega_j) - y^\delta(\omega_j)|^2 \le \delta^2. \tag{10}$$

The regularized approximation $x^\delta_{\alpha,m}$ based on noisy data is given as

$$x^\delta_{\alpha,m} = (\alpha I + H^*_\Omega H_\Omega)^{-1} H^*_\Omega S_\Omega y^\delta. \tag{11}$$

It also has the form (8) with the coefficients that solve the system (9), where $y(\omega_j)$ is substituted for $y^\delta(\omega_j)$.

Recall now that in the regularization theory the concept of source conditions is used to quantify the accuracy of an approximate solution. According to this concept [11] the Moore-Penrose generalized solution $x = x^\dagger$ of the equation (2), assuming it exists, can always be represented in terms of an operator-valued non-decreasing index function φ such that $\varphi(0) = 0$ and

$$x^\dagger = \varphi(H^*H)v, \quad \|v\|_{L_{2,\infty}} \le \rho. \tag{12}$$

Moreover, as it is explained in [14], when considered collocated Tikhonov regularization, such as (6), it is natural to assume that in (12) the index function φ is operator monotone on an interval containing the spectra of operators H^*H, $H_\Omega^* H_\Omega$. Then from Proposition 2.22 [10] it follows that

$$\|\varphi(H^*H) - \varphi(H_\Omega^* H_\Omega)\|_{L_{2,\infty}\to L_{2,\infty}} \leq d\, \varphi\big(\|H^*H - H_\Omega^* H_\Omega\|_{L_{2,\infty}\to L_{2,\infty}}\big), \tag{13}$$

where the coefficient d depends only on φ. Moreover, there exists a φ-dependent coefficient χ_φ such that

$$\sup_{\lambda>0} \frac{\alpha}{\alpha+\lambda}\varphi(\lambda) \leq \chi_\varphi \varphi(\alpha). \tag{14}$$

Note that examples of operator monotone index functions include the functions $\varphi(\lambda) = \lambda^\nu$, $0 < \nu \leq 1$, $\varphi(\lambda) = \log^{-\mu}(1/\lambda)$, $\lambda \in (0,1)$, $\mu > 0$, which traditionally used in the regularization theory to describe the solution smoothness in terms of source conditions.

Now, using the argument similar to that in [14] we can prove the following statement

Theorem 1. *Assume that source condition (12) with an operator monotone index function is satisfied. Then*

$$\|x^\dagger - x_{\alpha,m}^\delta\|_{L_{2,\infty}} \leq \frac{\delta}{2\sqrt{\alpha}} + C\Big(\varphi(\alpha) + \varphi\left(\|H^*H - H_\Omega^* H_\Omega\|_{L_{2,\infty}\to L_{2,\infty}}\right)\Big),$$

where the coefficient C depends only on φ and ρ involved in the condition (12).

Proof. To estimate the error $\|x^\dagger - x_{\alpha,m}^\delta\|_{L_{2,\infty}}$, we write the decomposition

$$x^\dagger - x_{\alpha,m}^\delta = (x^\dagger - x_{\alpha,m}) + (x_{\alpha,m} - x_{\alpha,m}^\delta).$$

For the first difference we have

$$x^\dagger - x_{\alpha,m} = x^\dagger - (\alpha I + H_\Omega^* H_\Omega)^{-1} H_\Omega^* S_\Omega y =$$

$$= (I - (\alpha I + H_\Omega^* H_\Omega)^{-1} H_\Omega^* H_\Omega)\, x^\dagger = T_1 + T_2,$$

where

$$T_1 = (I - (\alpha I + H_\Omega^* H_\Omega)^{-1} H_\Omega^* H_\Omega)\, \varphi(H_\Omega^* H_\Omega) v,$$

$$T_2 = (I - (\alpha I + H_\Omega^* H_\Omega)^{-1} H_\Omega^* H_\Omega)\, (\varphi(H^*H) - \varphi(H_\Omega^* H_\Omega))\, v.$$

By virtue of (13), (14) we have

$$
\begin{aligned}
\|T_1\|_{L_{2,\infty}} &\le \| \left(I-(\alpha I+H_\Omega^*H_\Omega)^{-1}H_\Omega^*H_\Omega\right)\varphi(H_\Omega^*H_\Omega)\|_{L_{2,\infty}\to L_{2,\infty}}\|v\|_{L_{2,\infty}} \le \\
&\le \rho\sup_\lambda \tfrac{\alpha}{\alpha+\lambda}\varphi(\lambda) \le \chi_\varphi\rho\varphi(\alpha),
\end{aligned}
$$

$$
\begin{aligned}
\|T_2\|_{L_{2,\infty}} &\le \|I-(\alpha I+H_\Omega^*H_\Omega)^{-1}H_\Omega^*H_\Omega\|_{L_{2,\infty}\to L_{2,\infty}}\|\varphi(H^*H)-\varphi(H_\Omega^*H_\Omega)\|_{L_{2,\infty}\to L_{2,\infty}}\|v\|_{L_{2,\infty}} \\
&\le d\rho\sup_\lambda \tfrac{\alpha}{\alpha+\lambda}\varphi\left(\|H^*H-H_\Omega^*H_\Omega\|_{L_{2,\infty}\to L_{2,\infty}}\right) \le d\rho\varphi\left(\|H^*H-H_\Omega^*H_\Omega\|_{L_{2,\infty}\to L_{2,\infty}}\right).
\end{aligned}
$$

It remains to estimate the difference $x_{\alpha,m}-x^\delta_{\alpha,m}$. To this end, we use the polar decomposition (see, for example, [21, p.35])

$$
H_\Omega^* = |H_\Omega|U^*, \tag{15}
$$

where $|H_\Omega| = (H_\Omega^*H_\Omega)^{1/2} : L_{2,\infty}\to L_{2,\infty}$ is the operator module of H_Ω and $U^*:\mathbb{R}^m\to L_{2,\infty}$ is the partial isometry operator, $\|U^*\|_{\mathbb{R}^m\to L_{2,\infty}}\le 1$. Then (15) allows us to continue as follows

$$
\begin{aligned}
\|x_{\alpha,m}-x^\delta_{\alpha,m}\|_{L_{2,\infty}} &= \|(\alpha I+H_\Omega^*H_\Omega)^{-1}H_\Omega^*S_\Omega(y-y^\delta)\|_{L_{2,\infty}} \le \\
&\le \|(\alpha I+H_\Omega^*H_\Omega)^{-1}H_\Omega^*\|_{\mathbb{R}^m\to L_{2,\infty}}\|S_\Omega(y-y^\delta)\|_{\mathbb{R}^m} \le \\
&\le \|(\alpha I+H_\Omega^*H_\Omega)^{-1}|H_\Omega|\|_{L_{2,\infty}\to L_{2,\infty}}\|U^*\|_{\mathbb{R}^m\to L_{2,\infty}}\|S_\Omega(y-y^\delta)\|_{\mathbb{R}^m} \le \\
&\le \delta/(2\sqrt{\alpha}).
\end{aligned}
$$

Thus, we obtain the desired estimate. □

Note that the term $\varphi(\|H^*H-H_\Omega^*H_\Omega\|_{L_{2,\infty}\to L_{2,\infty}})$ of the error bound given by Theorem 1 reflects a contribution of the collocation of operator H to the total error. In the sequel it is assumed that

$$\|H^*H - H_\Omega^* H_\Omega\|_{L_{2,\infty}\to L_{2,\infty}} \le \alpha. \tag{16}$$

In view of Theorem 1 such assumption ensures that the error caused by the collocation of H does not dominate the regularization error.

To balance all components of the error bound of Theorem 1 we consider the function

$$\Theta(\lambda) = \sqrt{\lambda}\varphi(\lambda), \qquad \lambda \in [0, \|H\|^2_{L_{2,\infty}\to L_{2,\Omega}}].$$

Theorem 2. *Let $\alpha = \Theta^{-1}(\delta)$ and the relation (16) hold. Then under the conditions of Theorem 1 we have*

$$\|x^\dagger - x^\delta_{\alpha,m}\|_{L_{2,\infty}} = O\Big(\varphi\left(\Theta^{-1}(\delta)\right)\Big). \tag{17}$$

Proof. It is clear that

$$\delta/\sqrt{\Theta^{-1}(\delta)} = \varphi\left(\Theta^{-1}(\delta)\right).$$

Then, by virtue of Theorem 1 and (16), we obtain

$$\|x^\dagger - x^\delta_{\alpha,m}\|_{L_{2,\infty}} \le \frac{\varphi\left(\Theta^{-1}(\delta)\right)}{2} + 2C\varphi\left(\Theta^{-1}(\delta)\right) = O\Big(\varphi\left(\Theta^{-1}(\delta)\right)\Big).$$

□

Note that, as it follows, for example, from [12], for given noise level δ and the source condition (12) the order (17) cannot be improved in general.

From the above discussion it follows that the condition (16) is important for the performance of the regularized collocation. The fulfilment of (16) depends on the kernel (4) and the sampling operator S_Ω. The properties of the later one are encoded in the distribution of sampling points ω_j, $j = 1, 2, \ldots, m$, and the weights γ_j, $j = 1, 2, \ldots, m$, inducing the inner product $\langle \cdot, \cdot \rangle_{\mathbb{R}^m}$. We will interprete them as knots and coefficients of a numerical integration formula and adopt the following assumption.

Assumption 1. *Assume that the quadrature formula*

$$\int_{\omega_1}^{\omega_m} f(\omega)\ d\omega \quad \simeq \quad \sum_{j=1}^{m} \gamma_j f(\omega_j) \tag{18}$$

is exact for polynomials of the 1st degree and for any function f having continuous derivatives $f^{(r)}$ in $[\omega_1, \omega_m]$ it holds

$$\left| \int_{\omega_1}^{\omega_m} f(\omega)\ d\omega - \sum_{j=1}^{m} \gamma_j f(\omega_j) \right| \leq c_{r,\Omega} h^r \max_{\omega_1 \leq \omega \leq \omega_m} |f^{(r)}(\omega)|, \tag{19}$$

where the coefficient $c_{r,\Omega} > 0$ depends on r, Ω and

$$h = \sup_{\omega \in [\omega_1, \omega_m]} \min_{\omega_j} |\omega - \omega_j|$$

is the so-called grid norm.

Remark 1. *The simplest example of formulae (18) satisfying Assumption 1 is the trapezoidal formula (for $r = 2$). More sophisticated constructions can be found in [8].*

In the sequel we restrict ourselves to the equations (1) with the kernels $K(\omega, \tau)$ admitting the representation

$$K(\omega, \tau) = \omega^{1/2} \varepsilon_1(\tau) + \varepsilon_2(\omega, \tau), \tag{20}$$

where the functions ε_1 and ε_2 are such that

a) $(1+\tau)^{-\beta} \varepsilon_1(\tau) \in L_{2,\infty}$,

b) $\frac{\partial^i \varepsilon_2(\omega,\tau)}{\partial \omega^i}$, $\frac{\partial^i (\omega^{1/2} \varepsilon_2(\omega,\tau))}{\partial \omega^i}$, $i = \overline{0, r}$, are continuous on $[\omega_1, \omega_m] \times [0, \infty)$ and

$$\sup_{\omega,\tau} \left| \frac{\partial^i \varepsilon_2(\omega,\tau)}{\partial \omega^i} \right| < \infty, \quad \sup_{\omega,\tau} \left| \frac{\partial^i (\omega^{1/2} \varepsilon_2(\omega,\tau))}{\partial \omega^i} \right| < \infty.$$

In the next section we will show that the representation (20) is relevant in the context of DDT-problem motivating the present study.

Consider the following auxiliary functions

$$g_0(\tau) = (1+\tau)^{-\beta} \max_{\omega} \left| \frac{\partial^r (\omega^{1/2}\varepsilon_2(\omega,\tau))}{\partial \omega^r} \right|,$$

$$g_1(\omega) = \sum_{i=0}^{r} C_i^r \left| \frac{\partial^{r-i}\omega^{1/2}}{\partial \omega^{r-i}} \right| \left(\int_0^\infty \left| \frac{\partial^i \varepsilon_2(\omega,u)}{\partial \omega^i} (1+u)^{-\beta} \right|^2 du \right)^{1/2},$$

$$g_2(\omega,\tau) = \sum_{i=0}^{r} C_i^r \left| \frac{\partial^{r-i}\varepsilon_2(\omega,\tau)}{\partial \omega^{r-i}} \right| \left(\int_0^\infty \left| \frac{\partial^i \varepsilon_2(\omega,u)}{\partial \omega^i} (1+u)^{-\beta} \right|^2 du \right)^{1/2},$$

where C_i^r are binomial coefficients. It is clear that g_0, g_1, g_2 are continuous and bounded on $[\omega_1, \omega_m] \times [0, \infty)$. Moreover, it is easy to see that

$$(1+\tau)^{-\beta} \max_{\omega} g_2(\omega,\tau) \in L_{2,\infty}.$$

Theorem 3. *Let Assumption 1 and conditions a), b) be fulfilled. Then for operators defined by (3), (4), (20) the following bound holds*

$$\|H^*H - H_\Omega^* H_\Omega\|_{L_{2,\infty}\to L_{2,\infty}} \le \bar{c}c_{r,\Omega}h^r,$$

where

$$\bar{c}^2 = \|(1+\cdot)^{-\beta}\varepsilon_1\|^2_{L_{2,\infty}} \left(\|g_0\|^2_{L_{2,\infty}} + \max_{\omega} |g_1(\omega)|^2 \right) + \|(1+\cdot)^{-\beta} \max_{\omega} g_2(\omega,\cdot)\|^2_{L_{2,\infty}}.$$

Proof. First of all we find a presentation for $H^*H - H_\Omega^* H_\Omega$. Recall that

$$Hx(\omega) = KJ^*x(\omega) = \int_0^\infty K(\omega,u)(1+u)^{-\beta}x(u)\, du,$$

$$H^*g(\tau) = JK^*g(\tau) = (1+\tau)^{\beta}K^*g(\tau) = (1+\tau)^{-\beta} \int_{\omega_1}^{\omega_m} K(\omega,\tau)g(\omega)\, d\omega.$$

Then

$$H^*Hx(\tau) = H^* \left(\int_0^\infty K(\tau,u)(1+u)^{-\beta}x(u)\, du \right) =$$

$$= (1+\tau)^{-\beta} \int_{\omega_1}^{\omega_m} K(\omega,\tau) \int_0^\infty K(\omega,u)(1+u)^{-\beta}x(u)\, du\, d\omega.$$

On the other hand

$$H_\Omega x = \left(\int\limits_0^\infty K(\omega_i, u)(1+u)^{-\beta} x(u)\ du\right)_{i=1}^m,$$

$$H_\Omega^* g(\tau) = \sum_{j=1}^m \gamma_j g_j K(\omega_j, \tau)(1+\tau)^{-\beta},$$

$$H_\Omega^* H_\Omega x(\tau) = \sum_{j=1}^m \gamma_j K(\omega_j, \tau)(1+\tau)^{-\beta} \int\limits_0^\infty K(\omega_j, u)(1+u)^{-\beta} x(u)\ du.$$

Therefore,

$$(H^*H - H_\Omega^* H_\Omega)x(\tau) \ = (1+\tau)^{-\beta}\Big(\int\limits_{\omega_1}^{\omega_m} K(\omega,\tau)\int\limits_0^\infty K(\omega,u)(1+u)^{-\beta}x(u)\ du\ d\omega -$$

$$-\sum_{j=1}^m \gamma_j K(\omega_j,\tau)\int\limits_0^\infty K(\omega_j,u)(1+u)^{-\beta}x(u)\ du\Big).$$

To simplify the calculations we introduce the notation

$$F(\omega,\tau) = K(\omega,\tau)\int\limits_0^\infty K(\omega,u)(1+u)^{-\beta}x(u)\ du. \tag{21}$$

Then

$$\|H^*H - H_\Omega^* H_\Omega\|^2_{L_{2,\infty}\to L_{2,\infty}} = \int\limits_0^\infty (1+\tau)^{-2\beta}\left(\int\limits_{\omega_1}^{\omega_m} F(\omega,\tau)\ d\omega - \sum_{j=1}^m \gamma_j F(\omega_j,\tau)\right)^2 d\tau. \tag{22}$$

By virtue of (20), (21), we have

$$F(\omega,\tau) \ = \left(\omega^{1/2}\varepsilon_1(\tau) + \varepsilon_2(\omega,\tau)\right)\int\limits_0^\infty \left(\omega^{1/2}\varepsilon_1(u) + \varepsilon_2(\omega,u)\right)(1+u)^{-\beta}x(u)\ du =$$

$$= F_1(\omega,\tau) + F_2(\omega,\tau) + F_3(\omega,\tau) + F_4(\omega,\tau),$$

where

$$F_1(\omega,\tau) = \omega\varepsilon_1(\tau)\int_0^\infty \varepsilon_1(u)(1+u)^{-\beta}x(u)\ du,$$

$$F_2(\omega,\tau) = \omega^{1/2}\varepsilon_2(\omega,\tau)\int_0^\infty \varepsilon_1(u)(1+u)^{-\beta}x(u)\ du,$$

$$F_3(\omega,\tau) = \omega^{1/2}\varepsilon_1(\tau)\int\limits_0^\infty \varepsilon_2(\omega,u)(1+u)^{-\beta}x(u)\ du,$$

$$F_4(\omega,\tau) = \varepsilon_2(\omega,\tau)\int\limits_0^\infty \varepsilon_2(\omega,u)(1+u)^{-\beta}x(u)\ du.$$

Thus, we have

$$(H^*H - H^*_\Omega H_\Omega)x(\tau) = (1+\tau)^{-\beta}\sum_{l=1}^{4}\left(\int_{\omega_1}^{\omega_m} F_l(\omega,\tau)\ d\omega - \sum_{j=1}^{m}\gamma_j F_l(\omega_j,\tau)\right)$$

with

$$\int\limits_{\omega_1}^{\omega_m} F_1(\omega,\tau)\ d\omega - \sum_{j=1}^{m}\gamma_j F_1(\omega_j,\tau) =$$

$$\varepsilon_1(\tau)\int\limits_0^\infty \varepsilon_1(u)(1+u)^{-\beta}x(u)\ du\left(\int\limits_{\omega_1}^{\omega_m}\omega\ d\omega - \sum_{j=1}^{m}\gamma_j\omega_j\right),$$

$$\int\limits_{\omega_1}^{\omega_m} F_2(\omega,\tau)\ d\omega - \sum_{j=1}^{m}\gamma_j F_2(\omega_j,\tau) =$$

$$= \int_0^\infty \varepsilon_1(u)(1+u)^{-\beta}x(u)\ du\left(\int\limits_{\omega_1}^{\omega_m}\omega^{1/2}\varepsilon_2(\omega,\tau)\ d\omega - \sum_{j=1}^{m}\gamma_j\omega_j^{1/2}\varepsilon_2(\omega_j,\tau)\right),$$

$$\int\limits_{\omega_1}^{\omega_m} F_3(\omega,\tau)\ d\omega - \sum_{j=1}^{m}\gamma_j F_3(\omega_j,\tau) =$$

$$= \varepsilon_1(\tau)\Big(\int\limits_{\omega_1}^{\omega_m}\omega^{1/2}\int\limits_0^\infty \varepsilon_2(\omega,u)(1+u)^{-\beta}x(u)\ du\ d\omega -$$

$$-\sum_{j=1}^{m}\gamma_j\omega_j^{1/2}\int\limits_0^\infty \varepsilon_2(\omega_j,u)(1+u)^{-\beta}x(u)\ du\Big),$$

$$\int\limits_{\omega_1}^{\omega_m} F_4(\omega,\tau)\ d\omega - \sum\limits_{j=1}^{m} \gamma_j F_4(\omega_j,\tau) =$$

$$= \int\limits_{\omega_1}^{\omega_m} \varepsilon_2(\omega,\tau) \int\limits_{0}^{\infty} \varepsilon_2(\omega,u)(1+u)^{-\beta} x(u)\ du\ d\omega -$$

$$- \sum\limits_{j=1}^{m} \gamma_j \varepsilon_2(\omega_j,\tau) \int\limits_{0}^{\infty} \varepsilon_2(\omega_j,u)(1+u)^{-\beta} x(u)\ du.$$

Let us estimate all the above components. Since the quadrature formula (18) is exact for polynomials of the 1st degree, we have

$$\int_{\omega_1}^{\omega_m} F_1(\omega,\tau)\ d\omega - \sum_{j=1}^{m} \gamma_j F_1(\omega_j,\tau) = 0.$$

With the help of relation (19) and conditions *a), b)*, we find

$$\int_{\omega_1}^{\omega_m} F_2(\omega,\tau)\ d\omega - \sum_{j=1}^{m} \gamma_j F_2(\omega_j,\tau) \le c_{r,\Omega} h^r \times$$

$$\times \max_{\omega} \left| \frac{\partial^r (\omega^{1/2} \varepsilon_2(\omega,\tau))}{\partial \omega^r} \right| \|(1+\cdot)^{-\beta} \varepsilon_1\|^2_{L_{2,\infty}} \|x\|^2_{L_{2,\infty}}. \tag{23}$$

Further, taking into account relation (19) and conditions *a), b)*, we obtain

$$\int_{\omega_1}^{\omega_m} F_3(\omega,\tau)\ d\omega - \sum_{j=1}^{m} \gamma_j F_3(\omega_j,\tau) \le c_{r,\Omega} h^r |\varepsilon_1(\tau)| \times$$

$$\times \max_{\omega} \left| \frac{\partial^r \left(\omega^{1/2} \int_0^\infty \varepsilon_2(\omega,u)(1+u)^{-\beta} x(u) du\right)}{\partial \omega^r} \right| \le$$

$$\le c_{r,\Omega} h^r |\varepsilon_1(\tau)| \max_{\omega} g_1(\omega) \|x\|^2_{L_{2,\infty}}. \tag{24}$$

Finally, using relation (19) and condition *b)*, we get

$$\int_{\omega_1}^{\omega_m} F_4(\omega,\tau)\ d\omega - \sum_{j=1}^{m} \gamma_j F_4(\omega_j,\tau) \le c_{r,\Omega} h^r \times$$

$$\times \max_{\omega} \left| \frac{\partial^r \left(\varepsilon_2(\omega,\tau) \int_0^\infty \varepsilon_2(\omega,u)(1+u)^{-\beta} x(u) du\right)}{\partial \omega^r} \right| \le$$

$$\le c_{r,\Omega} h^r \max_{\omega} g_2(\omega,\tau) \|x\|^2_{L_{2,\infty}}. \tag{25}$$

Substituting (23)–(25) into formula (22) we arrive at the desired estimate. □

3 Application to DDT-problem

In this section we interpret the outcome of our analysis for the context of DDT-problem. To be specific, we consider equation (1) with the kernel

$$K(\omega,\tau) = \sqrt{i\omega\tau}\tanh\sqrt{i\omega\tau}, \tag{26}$$

corresponding to DDT-problem for blocking boundary conditions and planar symmetry [17]. By separating real and imaginary parts we obtain that

$$K(\omega,\tau) = K_1(\omega,\tau)\frac{\sqrt{2\omega\tau}}{2} + iK_2(\omega,\tau)\frac{\sqrt{2\omega\tau}}{2},$$

with

$$K_1(\omega,\tau) = \frac{\sinh\sqrt{2\omega\tau} - \sin\sqrt{2\omega\tau}}{\cosh\sqrt{2\omega\tau} + \cos\sqrt{2\omega\tau}},$$
$$K_2(\omega,\tau) = \frac{\sinh\sqrt{2\omega\tau} + \sin\sqrt{2\omega\tau}}{\cosh\sqrt{2\omega\tau} + \cos\sqrt{2\omega\tau}}.$$

Since we are interested in a real-valued solution $p(\tau)$ of (1), this equation can be reformulated into a system of two integral equations of the form (1) with the kernels $K = K_1$, $K = K_2$ and $y(\omega) = Y_1(\omega)$, $y(\omega) = Y_2(\omega)$, where Y_1, Y_2 are real and imaginary parts of $Y(\omega) = Y_1(\omega) + iY_2(\omega)$. Note that a similar situation takes place in DRT-problem, where one often uses only a part of the impedance data Y_1 or Y_2 to reconstruct the quantity of interest (see, e.g., [3, 2, 23]). Here, in the same spirit, we consider an equation (1) with $K = K_1$ and $y = Y_1$ corresponding to the real part of DDT-problem. Moreover, it is convenient for us to change the variables as $\sqrt{\tau} \to \tau$, such that we arrive at an equation with operator H of the form (3), (4) with

$$K(\omega,\tau) = \sqrt{2\omega}K_1(\omega,\tau^2)\tau^2, \quad x(\tau) = (1+\tau)^{\beta}p(\tau^2). \tag{27}$$

Taking into account the form of $K_1(\omega,\tau^2)$ we have the representation

$$K(\omega,\tau) = \frac{\sinh\tau\sqrt{2\omega} - \sin\tau\sqrt{2\omega}}{\cosh\tau\sqrt{2\omega} + \cos\tau\sqrt{2\omega}}\sqrt{2\omega}\tau^2 =$$

$$= \sqrt{2\omega}\tau^2\left(1 - \left[1 - \frac{\sinh\tau\sqrt{2\omega} - \sin\tau\sqrt{2\omega}}{\cosh\tau\sqrt{2\omega} + \cos\tau\sqrt{2\omega}}\right]\right) =$$

$$= \sqrt{2\omega}\tau^2\left(1 - \frac{\cosh\tau\sqrt{2\omega} - \sinh\tau\sqrt{2\omega} + \cos\tau\sqrt{2\omega} + \sin\tau\sqrt{2\omega}}{\cosh\tau\sqrt{2\omega} + \cos\tau\sqrt{2\omega}}\right) =$$

$$= \sqrt{2\omega}\tau^2\left(1 - \frac{\mathrm{e}^{-\tau\sqrt{2\omega}} + \sqrt{2}\sin\left(\frac{\pi}{4} + \tau\sqrt{2\omega}\right)}{1 + \mathrm{e}^{-2\tau\sqrt{2\omega}} + 2\mathrm{e}^{-\tau\sqrt{2\omega}}\cos\tau\sqrt{2\omega}}2\mathrm{e}^{-\tau\sqrt{2\omega}}\right),$$

telling us that $K(\omega,\tau)$ has the form (20), where

$$\varepsilon_1(\tau) = \sqrt{2}\tau^2,$$

$$\varepsilon_2(\omega,\tau) = \sqrt{2\omega}\tau^2\frac{\mathrm{e}^{-\tau\sqrt{2\omega}} + \sqrt{2}\sin\left(\frac{\pi}{4} + \tau\sqrt{2\omega}\right)}{1 + \mathrm{e}^{-2\tau\sqrt{2\omega}} + 2\mathrm{e}^{-\tau\sqrt{2\omega}}\cos\tau\sqrt{2\omega}}2\mathrm{e}^{-\tau\sqrt{2\omega}}.$$

So, $\varepsilon_1(\tau)$ increases with $\tau \to \infty$ as $O(\tau^2)$. Therefore, the multiplier $p(\tau^2)$ of the unknown $x(\tau)$ should belong to L_2^β, $\beta > \frac{5}{2}$ to compensate such an increase. Therefore condition *a)* is satisfied with $\beta > 5/2$. Conditions *b)* can be easily verified directly.

To find a regularized solution of the integral equation (3), (4) with the above determined kernel we have the following system of linear algebraic equations, due to (9):

$$\alpha x_\alpha^j + \gamma_j\sum_{l=1}^{m} a_{jl}x_\alpha^l = \gamma_j y(\omega_j), \quad j = \overline{1,m} \tag{28}$$

with

$$a_{jl} = \sqrt{\omega_j\omega_l}\int_0^\infty \left[1 - Q(\omega_j,v)\right]\left[1 - Q(\omega_l,v)\right] v^4(1+v)^{-2\beta}\,dv,$$

where

$$Q(\omega_j,v) = \frac{\mathrm{e}^{-v\sqrt{2\omega}} + \sqrt{2}\sin\left(\frac{\pi}{4} + v\sqrt{2\omega}\right)}{1 + \mathrm{e}^{-2v\sqrt{2\omega}} + 2\mathrm{e}^{-v\sqrt{2\omega}}\cos v\sqrt{2\omega}}2\mathrm{e}^{-v\sqrt{2\omega}}.$$

For the above integrand we have

$$\left[1 - Q(\omega_j,v)\right]\left[1 - Q(\omega_l,v)\right] v^4(1+v)^{-2\beta} = q_c(v) + q_j(v) + q_l(v) + q_{jl}(v),$$

where

$$q_c(v) = \frac{v^4}{(1+v)^{2\beta}}, \quad q_j(v) = \frac{v^4}{(1+v)^{2\beta}} Q(\omega_j, v), \quad q_{jl}(v) = \frac{v^4}{(1+v)^{2\beta}} Q(\omega_j, v) Q(\omega_l, v),$$

$$j, l = \overline{1, m}.$$

It is clear that

$$I_1 = \int_0^\infty q_c(v) dv = \frac{24}{(2\beta-5)(2\beta-4)(2\beta-3)(2\beta-2)(2\beta-1)}. \tag{29}$$

To estimate the integrals of other components q_j, q_{jl} we recall [18] that if $f(x)$ is such that

$$|f(z)| \leq C \left(\frac{|z|}{1+|z|} \right)^\alpha \mathrm{e}^{-\beta \mathrm{Re}(z)}, \quad 0 < \alpha \leq 1, \quad 0 < \beta \leq 1, \tag{30}$$

for

$$z \in \{ z \in \mathbb{C} : \ |\arg(\sinh z)| < d \}, \quad 0 < d < \pi,$$

then (see e.g. [18] Example 4.2.11, p. 194 and Corollary 4.2.7, p. 188-189)

$$\begin{aligned} I(f) = \int_0^\infty f(x) dx \approx I_N(f) = h \sum_{k=-N}^{N} \frac{f(x_k)}{\sqrt{1+\mathrm{e}^{-2kh}}}, \\ x_k = \ln(\mathrm{e}^{kh} + \sqrt{1+\mathrm{e}^{2kh}}), \quad h = \frac{1}{\sqrt{N}}, \end{aligned}$$

(31)span

and there exist positive constants C, γ such that

$$|I(f) - I_N(f)| \leq C \exp(-\gamma \sqrt{N}).$$

One can check that the functions $q_j(x)$, $q_{jl}(x)$ satisfy the above condition (30). For example, for real values $z > 0$ we have

$$|q_j(z)| \leq 2 \frac{\left| \mathrm{e}^{-z\sqrt{2\omega_j}} + \sqrt{2} \sin\left(\frac{\pi}{4} + z\sqrt{2\omega_j}\right) \right|}{\left| 1 + \mathrm{e}^{-2z\sqrt{2\omega_j}} + 2\mathrm{e}^{-z\sqrt{2\omega_j}} \cos z\sqrt{2\omega_j} \right|} \mathrm{e}^{-z\sqrt{2\omega_j}} \frac{z^4}{(1+z)^{2\beta}} \leq$$

$$\leq \frac{2(1+\sqrt{2})}{0.87} \mathrm{e}^{-z\sqrt{2\omega_j}} \frac{z^4}{(1+z)^{2\beta}} \leq 5.55 \mathrm{e}^{-z\sqrt{2\omega_j}} \frac{z^4}{(1+z)^{2\beta}},$$

that corresponds to (30) with $\alpha = \beta = 1$. Similar bounds are valid for $q_{jl}(v)$.

Thus, using (29), (31) we estimate the coefficients of the system (28) as follows:

$$a_{jl} \approx I_1 + I_N(q_j) + I_N(q_l) + I_N(q_{jl}).$$

After solving the system (28), the regularized solution of the integral equation (3), (4) is defined by (8).

The next step is to select a value of the regularization parameter α to provide an accurate approximation for the solution of the problem (2)–(4), (27). The optimal value of α is defined in Theorem 2, but it requires a knowledge of the smoothness index function φ and the perturbation level δ. In the considered application such information is usually not available. To resolve this issue we propose to use the well-known quasi-optimality criterion [20], [6], [9]. Recall that in this criterion, we consider a geometric sequence of values of the regularization parameters $\alpha_n = \alpha_0 q^n$, $n = \overline{0, N}$, with a fixed $q < 1$ and $\alpha_0 > 0$. Then for each $\alpha = \alpha_n$ we compute the corresponding regularized approximation $x^\delta_{\alpha_{n,m}}$ given by (8), (28), and select the value $\alpha = \alpha_{n^*}$ with

$$n^* = \arg\min \left\{ \left\| x^\delta_{\alpha_{n,m}} - x^\delta_{\alpha_{n-1,m}} \right\|_{L_{2,\infty}}, \quad n = 1, 2, \ldots, N \right\}.$$

In the numerical tests below we use the quasi-optimality criterion with $\alpha_0 = 1$, $q = 0.5$, $N = 50$, and we stop to compute $x^\delta_{\alpha_{n,m}}$ at $n = k$ if

$$\left\| x^\delta_{\alpha_{k,m}} - x^\delta_{\alpha_{k-1,m}} \right\|_{L_{2,\infty}} > \left\| x^\delta_{\alpha_{k-1,m}} - x^\delta_{\alpha_{k-2,m}} \right\|_{L_{2,\infty}}.$$

4 Numerical experiments

We consider a DDT-problem formulated as the integral equation (1) with the kernel (26). First we simulate EIS data $y(\omega)$ in such a way that the exact solution of (1) is given as $p(\tau) = \exp(-\ln^2(\tau))/\tau$ (see Figure 1). Note that such $p(\tau)$ mimics some of DDT-functions considered in [17].

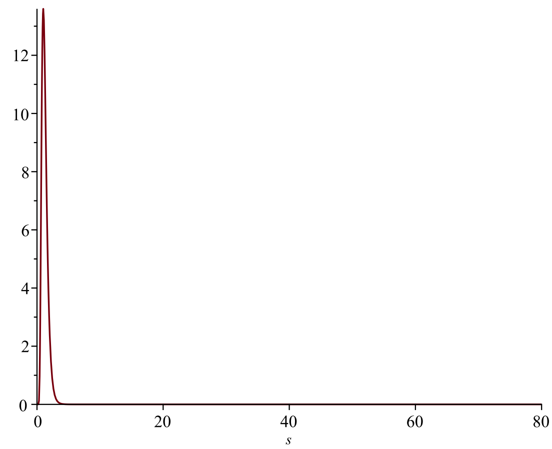


Figure 1: Synthetic DDT-function $p(\tau) = \exp(-\ln^2(\tau))/\tau$

EIS data sampling is simulated in the range $\Omega = [\omega_1, \omega_m]$ with $\omega_1 = 10^{-2}$, $\omega_m = 10^2$. Moreover, to be close to real EIS data set we suppose that sampling points ω_i are uniformly distributed in 4 decades $\Omega_j = [10^{j-2}, 10^{j-1}]$, $j = \overline{0,3}$, with 5 points in each decade. To illustrate the algorithm described in the previous section we apply it to the non-perturbed simulated data ($\delta = 0$, Example 1), and to noisy data $y^\delta(\omega_i) = y(\omega_i) + \delta_i$, $i = 1, 2, \ldots, m$, where δ_i are the realizations of zero-mean normally distributed random variables corresponding to noise-to-signal ratio of 1% (Example 2) and 10% (Example 3) respectively. The error observed in Example 1 is displayed in Figure 2. The reconstructions obtained for Examples 2 and 3 are displayed in Figures 3 and 4. The relative errors in $\|\cdot\|_{L_{2,\infty}}$-norm observed in these examples are 4.3% and 23% respectively. Keeping in mind the corresponding noise-to-signal ratios, one can conclude that in the considered examples the proposed algorithm demonstrates a stable and reliable behavior.

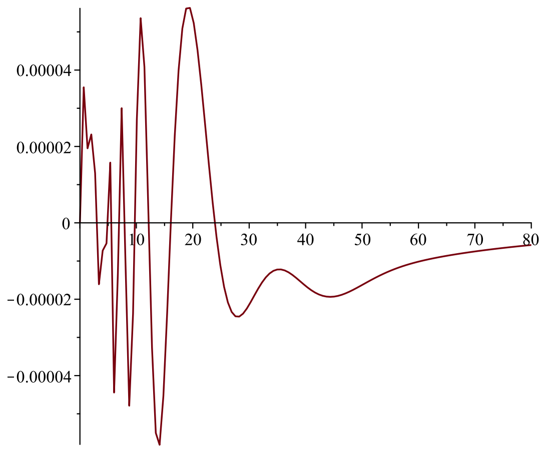


Figure 2: The error of the reconstruction from non-perturbed data: $\|p(\tau) - p_\alpha(\tau)\|_{L_{2,\infty}} = 0.581 * 10^{-4}$

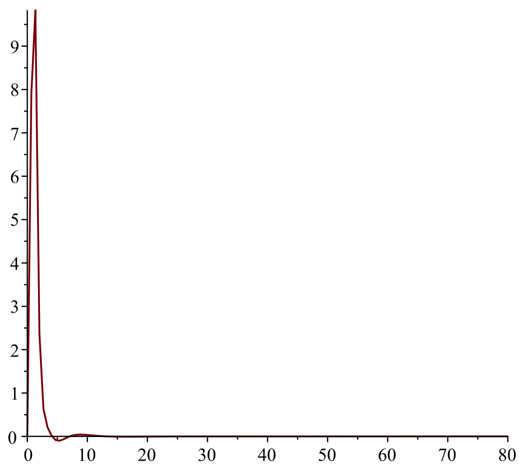


Figure 3: Example 2. Reconstruction from noisy data

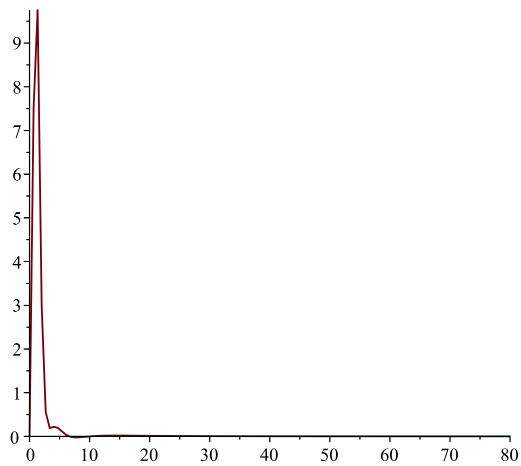


Figure 4: Example 3. Reconstruction from noisy data

We perform one more test, where EIS data $y(\omega)$ correspond to the exact solution of (1) given as $p(\tau) = \exp(-\ln(\tau)^2) + 1.3\exp(-2(2 - \ln(\tau))^2)$ (see Figure 5). This solution mimics a bimodal behavior of DDT-functions. Data simulations are performed in the same way as above. The algorithm is applied to non-perturbed data (Example 4) and to noisy data corresponding to noise-to-signal ratio of 2% (Example 5). The corresponding reconstructions are displayed in Figures 6 and 7.

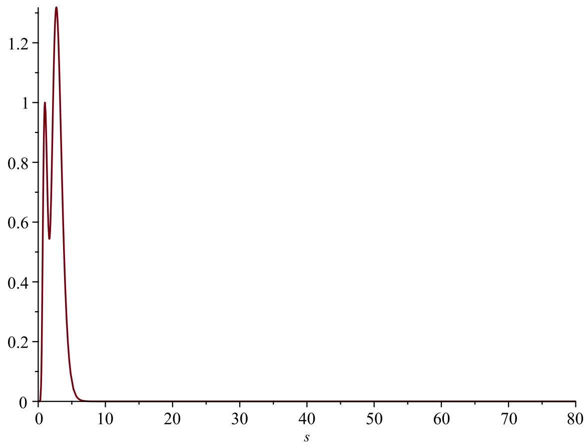
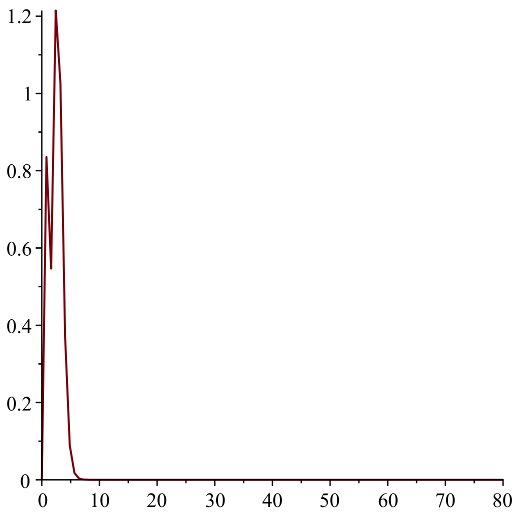


Figure 5: Bimodal synthetic DDT-function



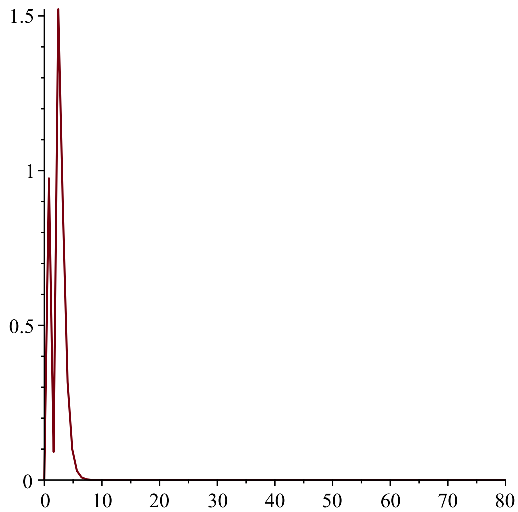


Figure 6: Example 4. Reconstruction from non-perturbed data

Figure 7: Example 5. Reconstruction from noisy data

Note that even in the case of non-perturbed data some reconstruction error unavoidably appears due to data discretization. In the considered case the discretization causes a relative error of 0.16%. The reconstruction from noisy data is given with a relative error of 5.2%. The results of this test demonstrate that the proposed algorithm is able to reconstruct important geometric features of DDT-functions such as the number of modes (picks) and their locations.

5 Conclusions

In this article we have analyzed a regularization scheme for solving Fredholm integral equations of the first kind with possibly non-square-integrable kernels. Such equations arise in EIS data processing performed in terms of DDT-functions. In our analysis we use a weighted space setting that allows an application of methods from the Regularization theory. The analyzed scheme is illustrated in application to DDT-problem for blocking boundary conditions and planar symmetry. In numerical implementation of the considered scheme we use fast (exponentially convergent) quadrature rules to find coefficients of a system of linear algebraic equations determining regularized solutions. Moreover, we use quasi-optimality criterion for selecting a value of the regularization parameter. Numerical experiments with synthetic data demonstrate reliability of the proposed scheme.

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