A. Bayesian deconvolution method

The relation between $\Delta W^{(e)}_{S,n}(\omega)$ and $\widetilde{\Delta W}^{(e)}_{S,n}(\omega)$ is given by the convolution product, see ~~C.(8-9)~~ (**??**) :

$$\widetilde{\Delta W}^{(e)}_{S,n}(\omega) = h_n(\omega) * \Delta W^{(e)}_{S,n}(\omega) = \int d\omega' h_n(\omega' - \omega)\Delta W^{(e)}_{S,n}(\omega') \tag{1}$$

In order to estimate $\Delta W^{(e)}_{S,n}(\omega)$ based on $\widetilde{\Delta W}^{(e)}_{S,n}(\omega)$, we need to implement a deconvolution algorithm. Since deconvolution is an ill-posed problem, simply performing a division in Fourier space leads to an estimation which is not robust to measurement errors. The sensibility to errors, due to lost informations, correspond to zero or close to zero values of the Fourier transform of h_n. In order to find a more robust estimation with correct physical properties, we propose to add appropriate prior information on $\Delta W^{(e)}_{S,n}(\omega)$ thanks to a Bayesian framework[49–51].

The discretized forward model for convolution (1) can be expressed as

$$\widetilde{\mathbf{\Delta W}}^{(e)}_{S,n} = \mathbf{H}_n.\mathbf{\Delta W}^{(e)}_{S,n} + \mathbf{N}_n, \tag{2}$$

where bold characters stand for vectors and matrices resulting from discretization. $\widetilde{\mathbf{\Delta W}}^{(e)}_{S,n}$ is the vector of data points, $\mathbf{H}_n$ is the convolution matrix, and $\mathbf{\Delta W}^{(e)}_{S,n}$ is the unknown quantity we are looking for. The term $\mathbf{N}_n$ is added to take account for all the errors (measurement and discretization). It is modeled as Gaussian random vector, with known covariance matrix $\mathbf{V}_e$ with diagonal elements $V_{e,i}$ estimated thanks to repeated experiments. This gives the expression of the probability distribution of $\widetilde{\mathbf{\Delta W}}^{(e)}_{S,n}$ knowing $\mathbf{\Delta W}^{(e)}_{S,n}$ and $\mathbf{V}_e$, which is called the likelihood :

$$p\left(\widetilde{\mathbf{\Delta W}}^{(e)}_{S,n}\middle|\mathbf{\Delta W}^{(e)}_{S,n}, \mathbf{V}_e\right) \propto \exp\left(-\frac{1}{2}\left|\left|\widetilde{\mathbf{\Delta W}}^{(e)}_{S,n} - \mathbf{H}_n.\mathbf{\Delta W}^{(e)}_{S,n}\right|\right|^2_{\mathbf{V}_e}\right) \tag{3}$$

where $||\mathbf{x}||^2_{\mathbf{V}_e} = \sum_i \frac{x_i^2}{v_{e_i}}$. Finding the argument which maximizes the likelihood, is equivalent to perform a division in Fourier space since the convolution matrix $\mathbf{H}_n$ is diagonal in the Fourier basis. This argument is dominated by $\mathbf{H}_n^{-1}\mathbf{N}_n$ terms.

In the Bayesian framework, by adding a prior information, we want to enforce physical properties such that $\Delta W^{(e)}_{S,n}(\omega)$ tends to zero when $|\omega|$ increases. For this purpose, we assign a Gaussian prior distribution on $\mathbf{\Delta W}^{(e)}_{S,n}$:

$$p\left(\mathbf{\Delta W}^{(e)}_{S,n}|\mathbf{V}_f\right) \propto \exp\left(-\frac{1}{2}\left|\left|\mathbf{\Delta W}^{(e)}_{S,n}\right|\right|^2_{\mathbf{V}_f}\right). \tag{4}$$

For variances $\mathbf{V}_f$, we use the expression :

$$V_f(\omega) = v_f \exp\left(-\frac{\omega^2}{w^2}\right), \tag{5}$$

where v_f and w are parameters tuned to enforce limit condition when $|\omega|$ increases.

Applying Bayes'rule, the posterior probability distribution of $\mathbf{\Delta W}^{(e)}_{S,n}$ combines likelihood (3) and prior distribution (4) :

$$p\left(\mathbf{\Delta W}^{(e)}_{S,n}|\widetilde{\mathbf{\Delta W}}^{(e)}_{S,n}, \mathbf{V}_e, \mathbf{V}_f\right) = \frac{p\left(\widetilde{\mathbf{\Delta W}}^{(e)}_{S,n}|\mathbf{\Delta W}^{(e)}_{S,n}, \mathbf{V}_e\right) p\left(\mathbf{\Delta W}^{(e)}_{S,n}|\mathbf{V}_f\right)}{p\left(\widetilde{\mathbf{\Delta W}}^{(e)}_{S,n}|\mathbf{V}_e, \mathbf{V}_f\right)}. \tag{6}$$

The argument which maximizes this posterior distribution (6), is the most likely estimate of $\mathbf{\Delta W}^{(e)}_{S,n}$ knowing both the measurement results $\widetilde{\mathbf{\Delta W}}^{(e)}_{S,n}$, $\mathbf{V}_e$, and prior information encoded in $\mathbf{V}_f$. Indeed, this Maximum A Posteriori (MAP), which also in this case is the Posterior Mean, is robust to errors $\mathbf{N}_n$. Because model evidence, the term in the denominator of (6), $p\left(\widetilde{\mathbf{\Delta W}}^{(e)}_{S,n}|\mathbf{V}_e, \mathbf{V}_f\right)$ does not depend on $\mathbf{\Delta W}^{(e)}_{S,n}$, MAP estimate becomes equivalent to the minimization of the criterion:

$$J(\mathbf{\Delta W}^{(e)}_{S,n}) = \frac{1}{2}\left|\left|\widetilde{\mathbf{\Delta W}}^{(e)}_{S,n} - \mathbf{H}_n.\mathbf{\Delta W}^{(e)}_{S,n}\right|\right|^2_{\mathbf{V}_e} + \frac{1}{2}\left|\left|\mathbf{\Delta W}^{(e)}_{S,n}\right|\right|^2_{\mathbf{V}_f}. \tag{7}$$

Estimated $\mathbf{\Delta W}^{(e)}_{S,n}$ has to comply with a box-constraint given by Pauli exclusion principle and Cauchy-Schwartz inequality[25] : implemented algorithm 1 looks for a minimum of criterion (7) inside the box-constraint thanks to a Projected Gradient Descent method[52]. In this algorithm, $M_n(\omega)$ denotes Cauchy-Schwartz inequality bounds, $\forall n, \omega$, and is explicitly given in Ref. 25.

In algorithm 1, $\mathbf{V}_f$ is assumed to be known thanks to expression (5). But in practice $\mathbf{V}_f$ is unknown, so it is estimated as well as $\mathbf{\Delta W}^{(e)}_{S,n}$. Then the estimate is both arguments $\mathbf{V}_f$ and $\mathbf{\Delta W}^{(e)}_{S,n}$ which maximize their joint posterior probability distribution. The method used to find the Joint Maximum A Posteriori (JMAP) as explained in Ref. 50 consists of assigning a conjugate Inverse Gamma prior distribution on $\mathbf{V}_f$ with shape and scale parameters α and $\beta(\omega)$, $\forall\omega$. Then obtaining the expression of the joint posterior probability of both unknowns and maximizing alternatively with respect to $\mathbf{\Delta W}^{(e)}_{S,n}$ as previously, and optimize the value of $\mathbf{V}_f$ with the following step:

$$V_f(\omega) = \frac{\beta(\omega) + \frac{1}{2}\Delta W^{(e)}_{S,n}(\omega)^2}{\alpha + \frac{3}{2}} \tag{8}$$

In this context, formula (5) is only used to initialize $\mathbf{V}_f$, β. The value of α determines the closeness of the optimized $\mathbf{V}_f$ to initialization (5). The closer α is to -1, the less $\mathbf{V}_f$ depends on initialization (5).

Algorithm 1 Detailed algorithm

Compute Cauchy-Schwartz bounds $M_n(\omega)$

Choose amplitude v_f and width w of prior (5) for $V_f(\omega)$

JMAP: initialize β with $\beta(\omega) = (\alpha + 1)V_f(\omega)$

Compute the minimum of criterion (7)

$\mathbf{\Delta W}_{S,n}^{(e)} = \left(\mathbf{H}^\top \mathbf{V}_e^{-1}\mathbf{H} + \mathbf{V}_f^{-1}\right)^{-1} \mathbf{H}^\top \mathbf{V}_e^{-1}\widetilde{\mathbf{\Delta W}}_{S,n}^{(e)}$

Project the solution inside the box given by Cauchy-Schwartz bounds:

$\Delta W_{S,n}^{(e)}(\omega) := \min(\Delta W_{S,n}^{(e)}(\omega), M_n(\omega))$

and $\Delta W_{S,n}^{(e)}(\omega) := \max(\Delta W_{S,n}^{(e)}(\omega), -M_n(\omega))$

repeat

Compute the gradient of criterion (7)

$\nabla\left(\mathbf{\Delta W}_{S,n}^{(e)}\right) = -\mathbf{H}^\top \mathbf{V}_e^{-1}\left(\widetilde{\mathbf{\Delta W}}_{S,n}^{(e)} - \mathbf{H}\mathbf{\Delta W}_{S,n}^{(e)}\right) + \mathbf{V}_f^{-1}\mathbf{\Delta W}_{S,n}^{(e)}$

Project the gradient $\mathbf{P}\nabla\left(\mathbf{\Delta W}_{S,n}^{(e)}\right)$ to stay in the box-constraint

if $|\Delta W_{S,n}^{(e)}(\omega)| \geq M_n(\omega)$ and $\Delta W_{S,n}^{(e)}(\omega) * \nabla\left(\Delta W_{S,n}^{(e)}\right)(\omega) \leq 0$ **then**

$\mathrm{P}\nabla\left(\Delta W_{S,n}^{(e)}\right)(\omega) = 0$

else

$\mathrm{P}\nabla\left(\Delta W_{S,n}^{(e)}\right)(\omega) = \nabla\left(\Delta W_{S,n}^{(e)}\right)(\omega)$

end if

Compute the furthest displacement d_∞ in the box along $\mathbf{P}\nabla\left(\mathbf{\Delta W}_{S,n}^{(e)}\right)$ direction

for all $\mathrm{P}\nabla\left(\Delta W_{S,n}^{(e)}\right)(\omega) \neq 0$ **do**

$d_\infty := \min\left(\frac{M_n(\omega) - \Delta W_{S,n}^{(e)}(\omega)}{\mathrm{P}\nabla\left(\Delta W_{S,n}^{(e)}\right)(\omega)}, d_\infty\right)$

end for

Compute the optimum displacement d_0 along $\mathbf{P}\nabla\left(\mathbf{\Delta W}_{S,n}^{(e)}\right)$ direction

$d_0 = \left|\left|\mathbf{P}\nabla\left(\mathbf{\Delta W}_{S,n}^{(e)}\right)\right|\right|^{-2}\left(\left|\left|\mathbf{HP}\nabla\left(\mathbf{\Delta W}_{S,n}^{(e)}\right)\right|\right|_{\mathbf{V}_e}^2 + \left|\left|\mathbf{P}\nabla\left(\mathbf{\Delta W}_{S,n}^{(e)}\right)\right|\right|_{\mathbf{V}_f}^2\right)$

Compute one projected descent gradient step

$\mathbf{\Delta W}_{S,n}^{(e)} := \mathbf{\Delta W}_{S,n}^{(e)} - \min(d_0, d_\infty)\mathbf{P}\nabla\left(\mathbf{\Delta W}_{S,n}^{(e)}\right)$

JMAP: optimize $\mathbf{V}_f$

$V_f(\omega) := \frac{\beta(\omega) + \frac{1}{2}\Delta W_{S,n}^{(e)}(\omega)^2}{\alpha + \frac{3}{2}}$

until MAP: $-\ln\left(p\left(\mathbf{\Delta W}_{S,n}^{(e)}\middle|\widetilde{\mathbf{\Delta W}}_{S,n}^{(e)}, \mathbf{V}_e, \mathbf{V}_f\right)\right)$

or JMAP: $-\ln\left(p\left(\mathbf{\Delta W}_{S,n}^{(e)}, \mathbf{V}_f\middle|\widetilde{\mathbf{\Delta W}}_{S,n}^{(e)}, \mathbf{V}_e, \beta, \alpha\right)\right)$ is minimized

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