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The Multidimensional Filter Diagonalization Method

I. Theory and Numerical Implementation

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The theory and numerical aspects of the recently developed multidimensional version of the filter diagonalization method (FDM) are described in detail. FDM can construct various "ersatz" or "hybrid" spectra from multidimensional time signals. Spectral resolution is not limited by the time-frequency uncertainty principle in each separate frequency dimension, but rather by the total joint information content of the signal, i.e., $N_{\text{total}} = N_1 \times N_2 \times \cdots \times N_D$, where some of the interferometric dimensions do not have to be represented by more than a few (e.g., two) time increments. It is shown that FDM can be used to compute various reduced-dimensionality projections of a high-dimensional spectrum directly, i.e., avoiding construction of the latter. A subsequent paper (*J. Magn. Reson.* 144, 357–366 (2000)) is concerned with applications of the method to 2D, 3D, and 4D NMR experiments.

INTRODUCTION

The filter diagonalization method (FDM) was originally designed by Neuhauser (*1*) for iterative diagonalization of large matrices which arise in quantum dynamics calculations when using a time-dependent approach. Later it was substantially modified and improved in a similar framework (see, e.g., Ref. (*2*) and references therein). Most importantly for the present paper, Wall and Neuhauser realized (*3*) that the method could be reformulated and split into two independent steps, namely, generation of a quantum time correlation function and its spectral analysis (or *harmonic inversion*). In this new formulation FDM is suitable for spectral analysis of a general experimentally measured time signal, simply by ignoring the first step of signal generation. FDM was conceptually new and potentially very promising, but its implementation was numerically inefficient. In Ref. (*4*) Mandelshtam and Taylor reformulated FDM for the conventional problem of processing a time signal defined on an evenly spaced time grid and found a way to significantly improve its performance. FDM has since found many applications in diverse fields and in particular for processing NMR time signals (*5–11*). In Ref. (*7*) we gave a detailed and systematic presentation of 1D FDM to the NMR community, also proposing a number of applications specific to NMR. Extension of the 1D FDM version of Ref. (*4*) to a model 2D case was presented by Mandelshtam and Taylor (*12*) and used to treat several NMR experiments (*5, 6*). We have recently improved the method further by introducing the idea of "averaging" several FDM calculations and applied the improved algorithm to homonuclear 2D *J* spectra (*10, 11*). Independently, Neuhauser and co-workers (*8, 9*) presented a related version of 2D FDM and applied it to a conventional COSY experiment, computing an absolute-value presentation of the phase-sensitive data. In our view, applications of 2D FDM to such spectra with *direct product* peak patterns cannot generally lead to an enormous resolution enhancement (or an equivalent significant reduction of the necessary signal size) compared to conventional strategies that analyze the spectra by processing 1D slices of the 2D signal. The reason is simply that in the second dimension of, say, the COSY experiment, both the information content of the signal *and* its complexity (the number of parameters required to characterize it) are increased due to the large number of cross peaks and their 2D multiplet structure. As will become clear, in FDM it is advantageous to minimize the total number of signal peaks K, so that they are dominated by the total number of measured time points $N_{\text{total}} = N_1 \times N_2 \times \cdots \times N_D$. It is also advantageous to minimize the number of peaks with degenerate frequencies (those that would appear, for example, along a single trace in one of the indirect dimensions). In certain 2D NMR experiments, including HSQC and 2D *J* spectroscopy and their possible combinations in more than two dimensions, the number of peaks does not increase multiplicatively when new time dimensions are introduced, and the direct product patterns do not occur. These basic experiments are therefore favorable for processing by multidimensional FDM. In the subsequent paper (*13*), here referenced as Paper II, these experimental implementations are described, as well as application to a TOCSY experiment, in which the number of peaks *is* multiplicative. Paper II also shows how a higher dimensional experiment can be used to project out a simpler spectrum.

The quantum mechanical language and notation used previously to derive FDM (*3, 4, 12*) is convenient and so are re-

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tained in the present paper with some modifications and adaptations for the multidimensional case. In the Appendix we give a summary of the notation and define the symbols we use. For the sake of brevity, we often assume that the reader is familiar with our previous papers (*4*, *7*) on 1D FDM.

We start by introducing a general complex valued D-dimensional time signal $c(\vec{n}) \equiv c(n_1\tau_1, n_2\tau_2, \ldots, n_D\tau_D)$, where $\vec{n}$ is the time vector, defined on an equidistant time grid. We will also call these time vectors *natural times.*

The total number of the natural time points, $N_{\text{total}} = N_1 \times N_2 \times \cdots \times N_D$, is only limited by instrument time and the computer disk capacity and does not usually exceed a gigabyte of data. While the number of the acquisition time points N_1 (we use this convention as it is more convenient for multidimensional signals) may be very large (say, of order of $N_1 \sim 10^3$–10^4), the number of time points in each of the other dimensions is strictly limited by the total experiment time and so is usually far less.

It is crucial to note that the spectral resolution of the conventional signal processing methods based on sequential applications of Fourier transform (FT) to the 1D slices of the D-dimensional signal is limited by the FT uncertainty principle in each dimension, i.e.,

$$\delta F_l \sim \frac{1}{N_l \tau_l}. \qquad [1]$$

In addition, an absorption-mode spectrum is always desired, which in the 1D case is obtained by simply taking the real part of the FT spectrum after it is correctly phased; in 2D NMR purely phase-modulated signals give rise to mixed-phase ("phase-twist") lineshapes in which neither the real nor the imaginary part of a 2D FT spectrum can be phased to the desired double-absorption lineshape (*14*). Absorption-mode spectra can be obtained from a pair of amplitude-modulated signals or from a pair of N- and P-type data sets by taking appropriate linear combinations (*15*). This necessitates using data sets twice as large. In 3D NMR the triple-absorption lineshape is obtained using by 2^2 bigger sets and so on. Note though that in some experiments, as in 2D *J*, the hypercomplex signals are unavailable, so that only absolute-value spectra can be obtained. The skew 1D 45° projection of a 2D absolute-value *J* spectrum (*16*), for example, leads to very poor resolution. Even if the FT in one or two dimensions is replaced by linear prediction (LP) followed by FT of the extended 1D signals, usually the situation does not change dramatically. With these conditions a 4D experiment is extremely expensive while a 5D NMR spectrum remains a thing of fantasy, as the total signal size must be unfeasibly huge to achieve even coarse frequency resolution.

In this paper we describe how various highly resolved, reduced-dimensionality, absorption-mode spectral projections of the multidimensional spectra can be obtained from purely phase-modulated signals. As demonstrated in Paper II under certain conditions such projections may need only a few (e.g., two) time points in some of the interferometric dimensions. This compression is achieved by applying FDM to the fully integrated, i.e., multidimensional harmonic inversion problem (HIP).

WHAT IS HIP?

A fully integrated D-dimensional HIP can be defined as the parametric fit of the full D-dimensional data set $c(\vec{n})$,

$$c(\vec{n}) = \sum_{k=1}^{K} d_k e^{-i\vec{n}\vec{\omega}_k} \equiv \sum_{k=1}^{K} d_k \exp[-i \sum_{l=1}^{D} n_l \tau_l \omega_{lk}], \qquad [2]$$

where $\vec{\omega}_k \equiv (\omega_{1k}, \omega_{2k}, \ldots, \omega_{Dk})$ are vectors of unknown complex frequencies, $\omega_{lk} = 2\pi f_{lk} - i\gamma_{lk}$, and d_k, unknown complex amplitudes. We will often refer to a pair $(\vec{\omega}_k, d_k)$ as a *spectral pole* and the set $\{\vec{\omega}_k, d_k\}$ as a *line list.* The total number of unknown complex parameters in the line list with K spectral poles is $(D + 1)K$. This formulation of HIP is similar to those proposed by others for a 2D spectral analysis (*17–20*). In the latter case the authors used models with a *direct product* set of frequencies, $\{\omega_{1k}, \omega_{2k'}, d_{kk'}\}$, $k = 1, 2, \ldots, K_1$, $k' = 1, 2, \ldots, K_2$, so that in the 2D plane the unknown spectral features would form a rectangular $K_1 \times K_2$ grid with total $K_1 + K_2 + K_1K_2$ number of unknowns. However, as will become clear later, this model with certain constraints is very similar to Eq. [2] with $D = 2$. Although it is convenient to start the presentation of multidimensional FDM with Eq. [2], for reasons explained below, in the numerical implementation of FDM to noisy data, we need not construct the line list itself to obtain the spectrum, so the actual form of the HIP, used only as a reference, will be irrelevant.

Generally speaking, Eq. [2] corresponds to a nonlinear optimization problem with totally $(D + 1)K$ complex fitting parameters. Because of this nonlinearity the exact solution of Eq. [2] may not exist at all. Even worse, an approximate solution might not be unique. As such, even for small data sets, solving Eq. [2] might be a very challenging project. Fortunately, Eq. [2] can be recast as an eigenvalue problem (or more precisely, as a family of generalized eigenvalue problems) partially avoiding these potential difficulties. In particular, existence and uniqueness of the solution for an eigenvalue problem are usually guaranteed.

If the HIP, Eq. [2], were to be solved by making a *global fit* of $c(\vec{n})$, the parameter K would be quite important to know in advance in order to avoid dealing with unnecessarily large and ill-conditioned matrices (*overfit*), on one hand, or solving a sequence of HIPs with ever increasing K, starting with a small K (*underfit*) until the result converges, on the other hand. This is often the case when high-resolution methods (e.g., LP) are used. In multidimensional FDM, just as in the 1D case (*4*, *7*), *the knowledge of K is irrelevant,* as the spectral analysis is

performed locally, i.e., for a given small spectral domain, so K will never appear in the formulas below. Instead, the total information content of the signal will unambiguously define the average density of spectral features used in the local spectral analysis.

SPECTRAL REPRESENTATIONS OF THE LINE LIST

In principle, the line list contains all of the information about the signal. The problem only is that such information given in a tabular format is often hard to absorb by a visually oriented operator, especially if there are too many overlapping spectral poles. Even in the 1D case we found it essential to compliment the line list with various types of "ersatz" spectra (*7*).

In the 1D case a converged FT spectrum should coincide, except for the noise background, with a converged FDM ersatz spectrum generated directly from the line list,

$$I(F) = \sum_k \frac{d_k}{\omega_k - i\Gamma - 2\pi F}, \qquad [3]$$

$$A(F) = \mathrm{Im}\{I(F)\}, \qquad [4]$$

where $I(F)$ and $A(F)$ stand, respectively, for complex and absorption spectra. The former is assumed to be phased correctly. The smoothing parameter Γ is often useful to improve the appearance of the spectra with very narrow lines. Such a smoothing is equivalent to an increase of the widths of all the Lorentzians by Γ. In the formulas below Γ is not shown although its implementation is always obvious.

The conventional 1D NMR FIDs decay with time and therefore it is usually assumed that all ω_k have negative imaginary parts. However, some complex frequencies obtained from the fit of a truncated signal may have positive imaginary parts due to either noise or imperfections of the fit. In such a case the "wrong" complex frequency is simply replaced by ω_k^* in Eq. [4] (see also the discussion in Ref. (*7*)).

Apparently, the analog of the conventional FT spectrum, Eq. [4], even when converged with respect to the length of the signal and even with an infinite signal to noise ratio (SNR), might not be the most revealing spectral representation, especially in cases of overlapping lines. However, the line list can be used to generate other types of ersatz spectra more suitable for a particular situation (*7*).

Under the assumption of Eq. [2] the D-dimensional complex FT of the purely phase-modulated signal is given by

$$I(\vec{F}) = \sum_k d_k \prod_{l=1}^{\mathrm{D}} \frac{1}{\omega_{lk} - 2\pi F_l}. \qquad [5]$$

Note though that unlike in the 1D case (Eq. [4]), a multidimensional absorption-mode spectrum cannot be obtained by taking either the real or the imaginary part of $I(\vec{F})$. Of course, Eq. [5] can (when, for instance, in a 2D experiment both N- and P-type signals are available) be used to produce an absorption-mode spectrum by taking certain combinations of the corresponding complex spectra. On the other hand, if a line list exists, construction of absorption-mode spectra is quite straightforward, even in the multidimensional case (*5*, *6*). One example of such a spectrum reads as

$$A(\vec{F}) = \sum_k \mathrm{Re}\{d_k\} \prod_{l=1}^{\mathrm{D}} \mathrm{Im}\left\{\frac{1}{\omega_{lk} - 2\pi F_l}\right\}, \qquad [6]$$

where $\vec{F} = (F_1, F_2, \ldots, F_{\mathrm{D}})$. Clearly, Eq. [6] is not the only possible representation. An alternative D-dimensional absorption spectrum, which has similar characteristics and is identical to Eq. [6] in the case of all real amplitudes d_k, may be written as

$$A(\vec{F}) = \sum_k \mathrm{Im}\left\{\frac{d_k}{\omega_{1k} - 2\pi F_1}\right\} \prod_{l=2}^{\mathrm{D}} \mathrm{Im}\left\{\frac{1}{\omega_{lk} - 2\pi F_l}\right\}. \qquad [7]$$

However, the two representations might differ significantly, e.g., result in different lineshapes, if numerically the amplitudes d_k are complex which happens in the cases of strongly overlapping and/or non-Lorentzian peaks. Note that any phase corrections, whether constant or frequency-dependent, must be applied to the complex amplitudes d_k before either formula can be used.

For the case of $\mathrm{D} > 2$ Eqs. [5]–[7] are useful but tedious to display: usually 1D or 2D projections or cross sections of multidimensional spectra are plotted. As such we introduce complex and absorption-mode 1D $\vec{p}$*-projections* along a time vector $\vec{p} = (p_1\tau_1, p_2\tau_2, \ldots, p_{\mathrm{D}}\tau_{\mathrm{D}})$ (*10*),

$$I_{\vec{p}}(F) = \sum_k \left\{\frac{d_k}{\omega_{\vec{p}k} - 2\pi F}\right\}, \qquad [8]$$

$$A_{\vec{p}}(F) = \mathrm{Im}\{I_{\vec{p}}(F)\}, \qquad [9]$$

where the $\vec{p}$-projections of the frequency vectors are

$$\omega_{\vec{p}k} = \frac{\vec{p}\,\vec{\omega}_k}{\tau_1} \equiv \frac{1}{\tau_1} \sum_l p_l \tau_l \omega_{lk}. \qquad [10]$$

Some important examples of $\vec{p}$-projections in the 2D case are the trivial projections corresponding to $\vec{p} = (0, \tau_2)$ or $\vec{p} = (\tau_1, 0)$ and the nontrivial 45° projection (*10*), $\vec{p} = (\tau_1, -\tau_1)$. Note that there is no FT analog for the latter in the case of purely phase-modulated signals. Other reduced dimensionality spectral projections are also possible, e.g., various 2D projections of 3D or 4D spectra, examples of which for real NMR data, namely, *singlet*-HSQC and *singlet*-TOCSY, are presented in Paper II.

In the line list of an FDM projection, the theoretical frequencies $\omega_{\vec{p}k} \equiv 2\pi f_{\vec{p}k} - i\gamma_{\vec{p}k}$ do not necessarily have all negative or all positive imaginary parts as the signal does not have to either decay or increase along the direction $\vec{p}$. While under such conditions an FT diverges (or in the case of a 2D J spectrum is exactly zero), the ersatz spectrum, Eq. [8] (with all negative $\gamma_{\vec{p}k}$ replaced by $-\gamma_{\vec{p}k}$ and, possibly with additional smoothing), exists and can provide very useful information.

In our previous papers (*5, 6, 12*) we demonstrated for some 2D cases that if the signal is not too noisy, i.e., the Lorentzian assumption of Eq. [2] holds, the line list is well defined and can be extracted from the signal. If this is not the case, solution of Eq. [2] simultaneously for *all* of the frequency components may not be accurate or even well defined. Rigorously speaking, only a 1D projection of the line list, i.e., a set $\{d_k, \omega_{\vec{p}k}\}$, can be both defined uniquely and computed to high precision using small data sets (*10*). However, we will see later that construction of well-resolved double-absorption ersatz spectra is also possible without explicitly using the line list, i.e., avoiding the use of Eqs. [5]–[7].

QUANTUM MECHANICAL ANSATZ TO SOLVE THE HIP: THE IDEAL CASE OF A NOISELESS SIGNAL

In this section we will show how to recast Eq. [2] as a generalized eigenvalue problem. We will essentially follow the derivation of Ref. (*12*), which, in turn, is a 2D extension of 1D FDM (*3, 4*).

We start with the conceptually simplest (albeit not general) case when the HIP assumption, Eq. [2], is exact for some finite number of poles. In this case it is possible to solve the HIP, Eq. [2], exactly and uniquely for the line list $\{d_k, \vec{\omega}_k\}$. Consequently, for the spectral reconstruction it is possible to use Eqs. [5]–[7].

Consider an operator vector $\vec{\Omega} = (\hat{\Omega}_1, \hat{\Omega}_2, \ldots, \hat{\Omega}_D)$ corresponding to a set of D *commuting non-Hermitian but symmetric* operators (Hamiltonians) with eigenvalues that coincide with the unknown frequencies ω_{lk}. For simplicity we assume that we can use the same set of eigenvectors Υ_k,

$$\hat{\Omega}_l \Upsilon_k = \omega_{lk} \Upsilon_k, \quad l = 1, 2, \ldots, D. \qquad [11]$$

(It will become clear later that this assumption is related to being able to construct a unique multidimensional line list, but creates certain conceptual and numerical difficulties.) The eigenvectors are orthonormalized,

$$(\Upsilon_k | \Upsilon_{k'}) = \delta_{kk'}, \qquad [12]$$

with respect to the complex symmetric inner product $(\phi|\psi) = (\psi|\phi)$. This actually implies the complex symmetric property of $\hat{\Omega}_l$,

$$(\Phi|\hat{\Omega}_l\Psi) = (\Psi|\hat{\Omega}_l\Phi) = (\hat{\Omega}_l\Psi|\Phi) \qquad [13]$$

for any two vectors Φ and Ψ. Note that the complex symmetric product does not define a norm, as it is generally complex valued, and therefore we cannot call the space formed by Υ_k a Hilbert space, a fact which fortunately has no direct bearing on the analysis.

We can now define a linear combination of the eigenvectors weighted with d_k as

$$\Phi(0) = \sum_k \sqrt{d_k} \Upsilon_k, \qquad [14]$$

here called the *initial state* for reasons that will become apparent later.

The Hamiltonian vector $\vec{\Omega}$ can be associated with the multitime *evolution operator,*

$$\hat{U}(\vec{n}) \equiv e^{-i\vec{n}\vec{\Omega}} \equiv \exp[-i \sum_{l=1}^{D} n_l \tau_l \hat{\Omega}_l]. \qquad [15]$$

With the above definitions we have constructed a quantum "dissipative" (because the evolution operators are not unitary) dynamical system. This quantum mechanical ansatz is a D-dimensional generalization of that invented by Wall and Neuhauser (*3*) for 1D FDM. The spectral properties of the underlying quantum system are implicitly defined by the spectral parameters, $\{\vec{\omega}_k, d_k\}$, of question. In particular, the time signal can now be written as a multitime quantum autocorrelation function:

$$c(\vec{n}) = (\Phi(0)|\hat{U}(\vec{n})\Phi(0)), \qquad [16]$$

which can be seen if we insert the spectral representation of the evolution operator,

$$\hat{U}(\vec{n}) = \sum_k e^{-i\vec{n}\vec{\omega}_k} |\Upsilon_k)(\Upsilon_k|, \qquad [17]$$

into Eq. [16] to recover Eq. [2]. This quantum ansatz will allow us to reformulate the HIP, Eq. [2], as a problem of diagonalizing, e.g., the basic evolution operators $\hat{U}_l = e^{-i\tau_l \hat{\Omega}_l}$ whose spectra yield the line list.

In the most general case, for two arbitrary time vectors $\vec{p} = (p_1\tau_1, p_2\tau_2, \ldots, p_D\tau_D)$ and $\vec{q} = (q_1\tau_1, q_2\tau_2, \ldots, q_D\tau_D)$ we can write a generalized eigenvalue equation

$$\hat{U}(\vec{p} + \vec{q})\Upsilon_k = u_k(\vec{p})\hat{U}(\vec{q})\Upsilon_k, \qquad [18]$$

where we used the property $\hat{U}(\vec{p} + \vec{q}) = \hat{U}(\vec{p})\hat{U}(\vec{q})$. According to Eq. [10] for the kth eigenvalue of $\hat{U}(\vec{p})$ we have

$$u_k(\vec{p}) \equiv e^{-i\vec{p}\vec{\omega}_k} \equiv e^{-i\tau_1\omega_{\vec{p}k}}. \qquad [19]$$

Solving Eq. [18] does not require the explicit knowledge of either $\hat{U}(\vec{p}+\vec{q})$ or $\hat{U}(\vec{q})$, once their matrix representations are available in a suitable basis. The most obvious choice for such a basis is

$$\Phi(\vec{n}) = \hat{U}(\vec{n})\Phi(0),\ n_l = 0,\ 1,\ \ldots,\ M_l, \qquad [20]$$

which we call the *Krylov basis*, because the vectors $\Phi(\vec{n})$ can also be represented as the result of repeated action of the operators $\hat{U}_l = e^{-i\tau_l\hat{\Omega}_l}$ on the initial state $\Phi(0)$. The total size of the Krylov basis is

$$M_{\text{Krylov}} = \prod_{l=1}^{D}(M_l+1). \qquad [21]$$

Assuming that it is sufficiently large to span the signal space, we can use it to expand a kth eigenvector,

$$\Upsilon_k = \sum_{\vec{n}} [\mathbf{B}_k]_{\vec{n}}\Phi(\vec{n}). \qquad [22]$$

Using the symmetry property of the evolution operator, which follows from Eq. [13] and Eq. [16] the matrix elements of $\hat{U}(\vec{p})$ between any two Krylov vectors are given by

$$\begin{aligned}[\mathbf{U}(\vec{p})]_{\vec{n}\vec{n}'} &\equiv (\Phi(\vec{n})|\hat{U}(\vec{p})\Phi(\vec{n}'))\\ &= (\hat{U}(\vec{n})\Phi(0)|\hat{U}(\vec{p})\hat{U}(\vec{n}')\Phi(0))\\ &= (\Phi(0)|\hat{U}(\vec{n}+\vec{n}'+\vec{p})\Phi(0))\\ &= c(\vec{n}+\vec{n}'+\vec{p}). \end{aligned} \qquad [23]$$

These matrix elements form a $M_{\text{Krylov}} \times M_{\text{Krylov}}$ complex symmetric matrix $\mathbf{U}(\vec{p})$ which can be computed by Eq. [23] only if the signal $c(\vec{n})$ is available at points with $p_l \le n_l \le 2M_l + p_l$.

We can now rewrite the eigenvalue problem, Eq. [18], in the matrix form,

$$\mathbf{U}(\vec{p}+\vec{q})\mathbf{B}_k = u_k(\vec{p})\mathbf{U}(\vec{q})\mathbf{B}_k, \qquad [24]$$

where due to Eqs. [12] and [22] the eigenvectors $\mathbf{B}_k$ (which are column vectors with elements $[\mathbf{B}_k]_{\vec{n}}$) are subject to orthonormalization with respect to the *overlap matrix* $\mathbf{U}_0 \equiv \mathbf{U}(0)$, i.e.,

$$\mathbf{B}_{k'}^{\mathrm{T}}\mathbf{U}_0\mathbf{B}_k = \delta_{kk'}. \qquad [25]$$

Due to Eqs. [14] and [22] the properly orthonormalized eigenvectors $\mathbf{B}_k$ can be used to compute the amplitudes,

$$\sqrt{d_k} = \sum_{\vec{n}} [\mathbf{B}_k]_{\vec{n}}c(\vec{n}). \qquad [26]$$

Note that implementation of Eq. [25] is trivial only if $\vec{q}=0$ or if the eigenvalue ω_{lk} is not degenerate, as in these cases $\mathbf{B}_{k'}^{\mathrm{T}}\mathbf{U}_0\mathbf{B}_k = 0$ automatically for $k \ne k'$ and one only has to normalize the kth eigenvector. This means that if there is no reason for $\vec{q}$ to be nonzero, e.g., if all the components $p_l \ge 0$, one has to choose $\vec{q}=0$.

For a general case, orthogonalization of the eigenvectors, i.e., solving Eq. [25], can be avoided by combining Eqs. [25] and [26] to yield

$$d_k = \sum_{k'} [\mathbf{S}^{-1}]_{kk'}b_k b_{k'}, \qquad [27]$$

with

$$\begin{aligned}[\mathbf{S}]_{kk'} &= \mathbf{B}_{k'}^{\mathrm{T}}\mathbf{U}_0\mathbf{B}_k,\\ b_k &= \sum_{\vec{n}} [\mathbf{B}_k]_{\vec{n}}c(\vec{n}),\end{aligned}$$

which will also result in correct amplitudes d_k. Note in addition that inversion of the matrix $\mathbf{S}$ is not necessary: to apply Eq. [27] one could solve the linear system,

$$\sum_k [\mathbf{S}]_{kk'}x_k = b_{k'} \qquad [28]$$

for x_k and then use the formula

$$d_k = x_k b_k. \qquad [29]$$

It appears that Eq. [23] can only be used with a natural time vector $\vec{p}$, i.e., corresponding to all integer values of p_l. However, as we showed in Ref. (*10*) if $\vec{p} = \alpha\vec{t}$ with $\vec{t}$ being natural, $\mathbf{U}(\vec{p})$ can be obtained by scaling the matrix $\mathbf{U}(\vec{t})$ as

$$\mathbf{U}(\vec{p}) \equiv \mathbf{U}(\alpha\vec{t}) = \sum_k (u_{\tilde{t}k})^{\alpha}\mathbf{U}_0\mathbf{B}_k\mathbf{B}_k^{\mathrm{T}}\mathbf{U}_0, \qquad [30]$$

where the eigenvalues $u_{\tilde{t}k}$ and the eigenvectors $\mathbf{B}_k$ are obtained by solving

$$\begin{aligned}\mathbf{U}(\vec{t})\mathbf{B}_k &= u_{\tilde{t}k}\mathbf{U}_0\mathbf{B}_k,\\ \mathbf{B}_k^{\mathrm{T}}\mathbf{U}_0\mathbf{B}_k &= 1\end{aligned} \qquad [31]$$

Some Remarks

Equation [30] together with Eqs. [23]–[29] have an interesting implementation in 2D *J* spectroscopy (*10*). Namely, if $\vec{q} = (0,\ \tau_1)$ (the case of $\vec{q} \ne 0$) and $\vec{p} = (\tau_1,\ -\tau_1)$ we have $\mathbf{U}(\vec{p}+\vec{q}) = \mathbf{U}(\tau_1,\ 0) \equiv \mathbf{U}_1$ and $\mathbf{U}(\vec{q}) = \mathbf{U}(0,\ \tau_1)$. The latter matrix can be obtained from $\mathbf{U}(0,\ \tau_2) \equiv \mathbf{U}_2$ using Eq. [30] with $\vec{t} = (0,\ \tau_2)$ and $\alpha = \tau_1/\tau_2$. The corresponding $\vec{p}$-projection leads to the broadband proton-decoupled proton NMR spectrum, in which all proton multiplets collapse to sharp singlets at the chemical shifts $\omega_{\vec{p}k}$. Note that for each collapsed multiplet $\omega_{\vec{p}k}$ will be degenerate so that the corresponding eigenvectors will not be automatically orthogonal, requiring the use of Eq. [27].

Given a projection vector $\vec{p}$, Eq. [24] defines a variational eigenvalue problem for $u_k(\vec{p})$ as well as for the frequency projections $\omega_{\vec{p}k}$. Notably, the amplitudes d_k *are not* obtained variationally, e.g., by solving a least-squares problem, and *do not* depend on the computed frequencies. This makes FDM quite different from all other methods of spectral analysis. For instance, the FFT corresponds to solving a variational problem for the amplitudes while fixing an equidistant frequency grid; LP methods solve a variational problem for the prediction coefficients which can in turn be used to find the spectral parameters by solving two more least-squares problems.

Equations [23]–[30] are working expressions and can, in principle, be used as are to obtain the estimates of the frequencies $\omega_{\vec{p}k}$ and amplitudes d_k for any D-dimensional signal and projection $\vec{p}$. Various numerical methods are available to solve the generalized eigenvalue problem, Eq. [24], our method of choice being the QZ algorithm (*21*) for complex general matrices, which is computationally competitive and accurate in the case of ill-conditioned or even singular matrices. However, the obvious technical difficulty, which is even worse than in the 1D case, is that the size, $M_{\text{Krylov}} \sim 2^{-\text{D}} \times N_{\text{total}}$, of the matrices involved is hardly feasible for any reasonably large multidimensional signal $c(\vec{n})$ with a total of $N_{\text{total}} = N_1 \times N_2 \times \cdots \times N_{\text{D}}$ data points. This difficulty is resolved in the next section, where a much more efficient basis is implemented to solve the eigenvalue problem.

A conceptual difficulty, which is not obvious, is associated with the construction of the complete line list $\{d_k, \vec{\omega}_k\}$, which implicitly requires the assumption that a $\mathbf{B}_k$ resulting from the solution of Eq. [24] for a particular $\vec{p}$, is simultaneously an eigenvector of Eq. [24] with any other $\vec{p}$ and $\vec{q}$. Only with the latter condition satisfied, at least approximately, can the different components ω_{lk} be "coupled" to form the eigenfrequency vector $\vec{\omega}_k \equiv (\omega_{1k}, \omega_{2k}, \ldots, \omega_{\text{D}k})$. Unfortunately, this is not generally the case unless either a model signal $c(\vec{n})$ exactly satisfying the form of Eq. [2] (*12*) or an experimental signal with high SNR (*5, 6*) is considered.

Another reason for $\mathbf{B}_k$ not being a unique eigenvector may be due to a degeneracy of the eigenvalue $u_k(\vec{p})$. This can easily occur in NMR spectra with cross peaks. Although, with high SNR, an otherwise degenerate spectrum still yields a unique 2D line list, for instance, by the simultaneous diagonalization method of Ref. (*9*), the latter would hardly become the method of choice in the case of low SNR.

We will revisit this problem in a subsequent section.

FOURIER BASIS

Our goal in this section is to solve Eq. [24] for a possibly very large data set $c(\vec{n})$. The solution of the problem is essentially analogous to that implemented in the 1D case (*3, 4, 7*). Namely, we want to introduce a *Fourier basis* along each "long" dimension n_l (where the large M_l causes the total Krylov basis to be large) by taking a small set of $K_{l\text{win}}$ equidistant frequencies φ_l within a chosen *frequency window* $[2\pi f_{l\min}, 2\pi f_{l\max}]$. Even though it is numerically more efficient to transform the Krylov basis only along the long dimensions, to make the appearance of the following expressions more compact, we consider Fourier transformation of the Krylov basis along all the D dimensions (the transformation along a "short" lth dimension being done with $K_{l\text{win}} = M_l + 1$ and the lth frequency window corresponding to the lth Nyquist range),

$$\begin{aligned}\Psi(\vec{\varphi}) &\equiv \Psi(\varphi_1, \varphi_2, \ldots, \varphi_{\text{D}}) \\ &= \sum_{\vec{n}} e^{i\vec{n}\vec{\varphi}}\Phi(\vec{n}) \\ &\equiv \sum_{n_1=0}^{M_1} e^{in_1\tau_1\varphi_1} \sum_{n_2=0}^{M_2} e^{in_2\tau_2\varphi_2} \ldots \sum_{n_{\text{D}}=0}^{M_{\text{D}}} e^{in_{\text{D}}\tau_{\text{D}}\varphi_{\text{D}}}\Phi(\vec{n}), \\ &\qquad 2\pi f_{l\min} < \varphi_l < 2\pi f_{l\max}. \end{aligned} \quad [32]$$

The resulting Fourier basis $\Psi(\vec{\varphi})$ in the D-dimensional window has size $K_{\text{win}} = K_{1\text{win}} \times K_{2\text{win}} \times \cdots \times K_{\text{Dwin}}$ and is much smaller than the original Krylov basis. As was argued for the 1D case (*3, 4, 7*), the Fourier subspace is only *locally complete*, i.e., it is "good" for a small subspace of eigenvectors corresponding to the chosen frequency window. That is, it is expected that those Υ_k, which are simultaneously the eigenvectors of the operators $\hat{\Omega}_l$ with eigenvalues $\omega_{lk} = 2\pi f_{lk} - i\gamma_{lk}$ satisfying $f_{l\min} < f_{lk} < f_{l\max}$, $l = 1, 2, \ldots, \text{D}$, can be expanded in the window basis as

$$\Upsilon_k = \sum_{\vec{\varphi}} [\mathbf{B}_k]_{\vec{\varphi}}\Psi(\vec{\varphi}), \quad [33]$$

where as in Eq. [32] the summation is carried out over $K_{\text{win}} = K_{1\text{win}} \times K_{2\text{win}} \times \cdots \times K_{\text{Dwin}}$ values of $\vec{\varphi}$. The number $K_{l\text{win}}$ of the frequency values φ_l in the lth window does not have to be an adjustable parameter of the method. It is usually chosen according to the *information content* of the finite signal $c(\vec{n})$ (i.e., its extension in the corresponding dimension) or that of the Krylov subspace $\{\Phi(\vec{n})\}$ (*4*),

$$K_{l\text{win}} = \rho(M_l + 1)(f_{l\max} - f_{l\min})\tau_l, \quad [34]$$

with ρ being usually chosen between 1 and 1.2. The maximum $K_{l\text{win}}$ is equal to $M_l + 1$ (the number of Krylov basis functions in the lth dimension) which would correspond to the maximum possible frequency interval, namely, the spectral width (Nyquist range), $\text{SW}_l = 1/\tau_l$. Note that for long dimensions the frequency interval is always chosen to be much less than the spectral width, so that $K_{l\text{win}} \ll M_l$. This means that we do not usually use the total information content of the signal, the price to pay for both the enormous reduction of the numerical effort and the necessity to make the algorithm numerically stable.

Implementation of the Fourier basis is analogous to that of Refs. (*4, 7*) and for the $\text{D} = 1$ case the resulting expressions are

equivalent. The corresponding matrix elements of $\hat{U}(\vec{p})$ are obtained by using the definition of the Fourier basis, Eq. [32], and the result of Eq. [23],

$$[\mathbf{U}(\vec{p})]_{\vec{\varphi}\vec{\varphi}'} \equiv (\Psi(\vec{\varphi})|\hat{U}(\vec{p})\Psi(\vec{\varphi}')) = \sum_{\vec{n}} \sum_{\vec{n}'} c(\vec{n}+\vec{n}'+\vec{p}) e^{i\vec{n}\vec{\varphi}} e^{i\vec{n}'\vec{\varphi}'}, \qquad [35]$$

where as in Eq. [32] the multiple summations are carried out over n_l, $n_l' = 0, 1, \ldots, M_l$ for each $l = 1, 2, \ldots, \mathrm{D}$.

Equation [35] is, in principle, a working expression, although quite expensive to evaluate numerically. Fortunately, we can get rid of half of the summations in Eq. [35] by first noticing that the double-Fourier transformation along each dimension is identical to that of the 1D signal (*4*, *7*):

$$U(\ldots, p_l, \ldots)_{(\ldots,\varphi_l,\ldots)(\ldots,\varphi_l',\ldots)} = \ldots \sum_{n_l=0}^{M_l} \sum_{n_l'=0}^{M_l} e^{in_l\tau_l\varphi_l} e^{in_l'\tau_l\varphi_l'} \ldots \times \ldots c(\ldots, n_l + n_l' + p_l, \ldots), \qquad [36]$$

where variables irrelevant to the lth dimension are suppressed. Now substituting $n_l \rightarrow n_l + n_l'$ and then evaluating the sum over n_l' we obtain

$$U(\ldots, p_l, \ldots)_{(\ldots,\varphi_l,\ldots)(\ldots,\varphi_l',\ldots)} = \ldots \hat{S}_l \sum_{\sigma_l=0,1} \frac{e^{i\sigma_l[\tau_l(M_l+1)(\varphi_l'-\varphi_l)+\pi]}}{1-e^{i\tau_l(\varphi_l'-\varphi_l)}} \times \sum_{n_l=\sigma_l(M_l+1)}^{(\sigma_l+1)M_l} e^{in_l\tau_l\varphi_l} \ldots c(\ldots, n_l + p_l, \ldots), \qquad [37]$$

where $\hat{S}_l$ defines the symmetrization operator over the variables φ_l, φ_l':

$$\hat{S}_l g(\varphi_l, \varphi_l') = g(\varphi_l, \varphi_l') + g(\varphi_l', \varphi_l). \qquad [38]$$

Equation [37] is correct for all choices of φ_l and φ_l', except for the singularity arising at $\varphi_l = \varphi_l'$. To obtain a numerically practical expression for this singular case we evaluate the $\varphi_l \rightarrow \varphi_l'$ limit leading to

$$U(\ldots, p_l, \ldots)_{(\ldots,\varphi_l,\ldots)(\ldots,\varphi_l,\ldots)} = \ldots \sum_{n_l=0}^{2M_l} e^{in_l\tau_l\varphi_l} \times (M_l - |M_l - n_l| + 1) \ldots c(\ldots, n_l + p_l, \ldots). \qquad [39]$$

The transformation along n_l is independent of the other indices, so implementation of Eqs. [37]–[39] is straightforward. As such for $\varphi_l \neq \varphi_l'$, $l = 1, 2, \ldots, \mathrm{D}$ we can write

$$[\mathbf{U}(\vec{p})]_{\vec{\varphi}\vec{\varphi}'} = \sum_{\sigma_1=0,1} \ldots \sum_{\sigma_\mathrm{D}=0,1} \times \left\{ \prod_{l=1}^{\mathrm{D}} \hat{S}_l \frac{e^{i\sigma_l[\tau_l(M_l+1)(\varphi_l'-\varphi_l)+\pi]}}{1-e^{i\tau_l(\varphi_l'-\varphi_l)}} \right\} \times \sum_{n_1=\sigma_1(M_1+1)}^{(\sigma_1+1)M_1} \ldots \sum_{n_\mathrm{D}=\sigma_\mathrm{D}(M_\mathrm{D}+1)}^{(\sigma_\mathrm{D}+1)/M_\mathrm{D}} e^{i\vec{n}\vec{\varphi}} c(\vec{n}+\vec{p}). \qquad [40]$$

As explained above, the singular cases are handled using Eq. [39]. For example, for $\varphi_1 = \varphi_1'$ Eq. [40] is rewritten as

$$[\mathbf{U}(\vec{p})]_{\vec{\varphi}\vec{\varphi}'} = \sum_{\sigma_2=0,1} \ldots \sum_{\sigma_\mathrm{D}=0,1} \times \left\{ \prod_{l=2}^{\mathrm{D}} \hat{S}_l \frac{e^{i\sigma_l[\tau_l(M_l+1)(\varphi_l'-\varphi_l)+\pi]}}{1-e^{i\tau_l(\varphi_l'-\varphi_l)}} \right\} \times \sum_{n_1=0}^{2M_1} (M_1 - |M_1 - n_1| + 1) \times \sum_{n_2=\sigma_2(M_2+1)}^{(\sigma_2+1)M_2} \ldots \sum_{n_\mathrm{D}=\sigma_\mathrm{D}(M_\mathrm{D}+1)}^{(\sigma_\mathrm{D}+1)M_\mathrm{D}} e^{i\vec{n}\vec{\varphi}} c(\vec{n}+\vec{p}), \qquad [41]$$

with similar expressions to treat other singularities. Finally for $\vec{\varphi} = \vec{\varphi}'$, i.e., the diagonal elements of the **U** matrix, we have

$$[\mathbf{U}(\vec{p})]_{\vec{\varphi}\vec{\varphi}} = \sum_{n_1=0}^{2M_1} (M_1 - |M_1 - n_1| + 1) \times \sum_{n_2=0}^{2M_2} (M_2 - |M_2 - n_2| + 1) \ldots \times \sum_{n_\mathrm{D}=0}^{2M_\mathrm{D}} (M_\mathrm{D} - |M_\mathrm{D} - n_\mathrm{D}| + 1) e^{i\vec{n}\vec{\varphi}} c(\vec{n}+\vec{p}), \qquad [42]$$

which is simply a D-dimensional FT of the signal $c(\vec{n}+\vec{p})$ with a symmetric triangular apodization function in each dimension.

Equation [32] defines the transformation from the Krylov vectors $\Phi(\vec{n})$ to the Fourier basis $\Psi(\vec{\varphi})$. According to this transformation the formula for the amplitudes, Eq. [26], is rewritten as

$$\sqrt{d_k} = \sum_{\vec{\varphi}} [\mathbf{B}_k]_{\vec{\varphi}} \sum_{n_1=0}^{M_1} \sum_{n_2=0}^{M_2} \ldots \sum_{n_\mathrm{D}=0}^{M_\mathrm{D}} e^{i\vec{n}\vec{\varphi}} c(\vec{n}), \qquad [43]$$

where the coefficients of the kth eigenvector, $[\mathbf{B}_k]_{\vec{\varphi}}$, resulting from Eq. [24] define the expansion of the kth eigenvector Υ_k in terms of the Fourier basis functions $\Psi(\vec{\varphi})$, Eq. [33].

To conclude this section we mention some obvious technical aspects of implementing Eqs. [40]–[43].

First, the multidimensional FTs of $c(\vec{n})$ should be carried out by appropriate FFT algorithms, which can be implemented using the globally equidistant grid of frequency points φ_l in each lth dimension.

Second, once FT of $c(\vec{n})$ is computed, FT of $c(\vec{n} + \vec{p})$ is obtained recursively. For example, this can be seen from the relationship

$$\sum_{n=0}^{M} e^{in\varphi} c(n+1) = [\sum_{n=0}^{M} e^{in\varphi} c(n) - c(0)]e^{-i\varphi} + c(M+1)e^{iM\varphi}.$$

Third, Eq. [43] is computationally very inexpensive as the corresponding Fourier sum has been already computed to evaluate $\mathbf{U}_0$ (see Eq. [40]). Note, however, that there exists a potentially more accurate expression for a coefficient d_k, which depends on the corresponding frequency vector $\vec{\omega}_k$ (*12*).

EXISTENCE AND UNIQUENESS OF THE SOLUTION

As we already mentioned, Eq. [2] corresponds to a nonlinear multiparameter optimization problem. Uniqueness or even existence of its solution is not always obvious and, in fact, for a general finite D-dimensional signal we can only show that a 1D $\vec{p}$-projection of the line list exists due to the existence of the corresponding solution of Eq. [24]. This is already a very important aspect as it allows us to obtain useful spectral information from the multidimensional signals which would not be available otherwise. Of course, the projected line list, i.e., $\{\omega_{\vec{p}k}, d_k\}$, $k = 1, 2, \ldots, K$, is still a function of the size of the signal. In particular, the total number of poles K that can be produced by FDM depends on the signal size, $N_{\text{total}} = N_1 \times N_2 \times \cdots \times N_{\text{D}}$. The reason simply is that, due to the noise always present in the experimental signal, it is never a particular finite sum of sinusoidal terms with fixed K as manifested by Eq. [2]. The number K is dictated by the information content of the finite signal. In the FDM framework K is limited by M_{Krylov} (see Eq. [21]), on one hand, but generally cannot be less than M_{Krylov} due to the noise. Thus for a general noisy signal we can safely write

$$K = M_{\text{Krylov}} \equiv 2^{-\text{D}} \times N_{\text{total}}. \qquad [44]$$

Equation [44] essentially eliminates the problem of choosing the "right" K, so typical in all other *high-resolution* methods based on a parametric fit.

It is useful to mention the case when the complete solution (in addition to the projections) is unique. If a signal does have exactly the form of Eq. [2] with particular fixed $K = K_0$ and zero noise, then for $M_{\text{Krylov}} \geq K$ the Krylov basis becomes complete and solving Eq. [24] with a suitable $\vec{p}$ should lead to a unique set of eigenvectors $\mathbf{B}_k$. These eigenvectors then diagonalize the matrix of any evolution operator, in particular, that of $\hat{U}_l = e^{-i\tau_l \hat{\Omega}_l}$. Therefore we have,

$$e^{-i\tau_l \omega_{lk}} = \frac{\mathbf{B}_k^{\text{T}} \mathbf{U}_l \mathbf{B}_k}{\mathbf{B}_k^{\text{T}} \mathbf{U}_0 \mathbf{B}_k}, \qquad [45]$$

which, in principle, yields the whole line list $\{\vec{\omega}_k, d_k\}$. Again, one has to be careful when dealing with degenerate frequencies ω_{lk}, typical for some NMR experiments like 2D COSY with $\vec{p} = (0, \tau_2)$ or $\vec{p} = (\tau_1, 0)$. In Ref. (*9*) the authors proposed a procedure called "simultaneous diagonalization," based on the Jacobi method, to overcome the degeneracy problem. However, such a degeneracy could be avoided by, e.g., avoiding the wrong choices for the projection vector $\vec{p}$ or by choosing small windows allowing only nondegenerate frequencies. Therefore degeneracy alone does not make the solution nonunique as long as Eq. [2] holds. Another question concerns the robustness of the FDM solution with respect to small variations of the input data (e.g., an experimental noise) that destroys the form of Eq. [2] corresponding to a finite K. It is definitely quite robust for 1D signals (*4*, *7*), where the problem of "coupling the frequency components" does not occur. For $\text{D} > 1$ it seems not to be the case. In practice, only a signal with high SNR that fits Eq. [2] well can be inverted to yield a reliable line list as we showed in Refs. (*5*, *6*, *12*). Note though that even with moderate noise Eq. [45], being *nonvariational*, is relatively inaccurate since the eigenvectors $\mathbf{B}_k$ are very sensitive to the small variations of the $\mathbf{U}$ matrices. In Refs. (*5*, *6*, *12*) a somewhat more sophisticated procedure was implemented to assign ω_{1k} to the right ω_{2k}, each computed variationally. This procedure was based on using several sets $\{u_k(\vec{p}), d_k\}$ with different $\vec{p}$ and the relation $u_k(\vec{p} + \vec{q}) = u_k(\vec{p})u_k(\vec{q})$. We do not describe this procedure in detail since it will also fail for very noisy signals and since we have found a better solution (*11*) described below.

For general noisy signals the assumption that the Hamiltonians $\hat{\Omega}_l$ commute does not imply that their finite matrix representations do, and so the eigenvector sets for different $\vec{p}$ may differ significantly. In such a case the simultaneous diagonalization based on the Jacobi method of Ref. (*9*) may be one way to attack the problem. However, there is no guarantee that this method converges at all, as it is essentially a nonlinear optimization of the kind we are trying to avoid by using only linear algebraic techniques to fit the data. In the next section we describe in detail another approach for dealing with the same problem which has proven to be quite reliable (*11*).

SPECTRAL RECONSTRUCTION USING GREEN'S FUNCTIONS: THE GENERAL CASE OF NOISY SIGNALS

1D Projections and Cross Sections

For a natural time vector $\vec{p}$ calculation of the projection is as simple as solving Eq. [24]. However, as we already discussed

above, if it is not one of the trivial projections corresponding to diagonalizing one of the basic evolution operators $\hat{U}_l$, a useful projection might not correspond to a natural $\vec{p}$ with all components p_l being both integer and small (to avoid the folding problem) as the time increments τ_l are often quite different.

One nontrivial and very useful example of a $\vec{p}$-projection is the 45° projection of a 2D J spectrum, in which the proton multiplets collapse to single peaks at the frequencies of the proton chemical shifts. As we demonstrated in Ref. (*10*) a highly resolved absorption-mode 45° projection can be obtained in the framework of FDM essentially using the approach outlined by Eqs. [23]–[30]. In the FFT framework construction of absorption-mode 45° projections is impossible, so only skew 45° projection of an absolute value 2D J spectrum (*16*) can be used.

In the following we generalize the procedure of calculating the 1D projections developed in Ref. (*10*) for the 2D J experiment to the case of arbitrary $\vec{p}$ and D. We slightly deviate from Ref. (*10*) and the above derivation, although conceptually the methods are similar.

First we introduce the $\vec{p}$-projection of the Hamiltonian vector in analogy with Eq. [10],

$$\hat{\Omega}_{\vec{p}} = \frac{1}{\tau_1}\vec{p}\vec{\Omega} \equiv \frac{1}{\tau_1}\sum_l p_l\tau_l\hat{\Omega}_l. \qquad [46]$$

Clearly we can write

$$\hat{\Omega}_{\vec{p}}\Upsilon_{\vec{p}k} = \omega_{\vec{p}k}\Upsilon_{\vec{p}k}, \qquad [47]$$

where for this general case we have also labeled the eigenfunctions of $\hat{\Omega}_{\vec{p}}$ with the additional subscript $\vec{p}$ emphasizing that they could be different for different Hamiltonians (due to possible degeneracies or noise), although we still assume that these Hamiltonians commute with each other.

We can now introduce a *Green's function* or *resolvent operator* $\hat{G}_{\vec{p}}(F)$ associated with $\hat{\Omega}_{\vec{p}}$ as

$$\hat{G}_{\vec{p}}(F) \equiv \frac{1}{\hat{\Omega}_{\vec{p}} - 2\pi F} = \sum_k \frac{|\Upsilon_{\vec{p}k})(\Upsilon_{\vec{p}k}|}{\omega_{\vec{p}k} - 2\pi F}, \qquad [48]$$

where we also used the spectral representation form assuming that the eigenfunctions $\Upsilon_{\vec{p}k}$ are orthonormalized as in Eq. [12].

Note that if we set $d_k = d_{\vec{p}k} = (\Phi(0)|\Upsilon_{\vec{p}k})^2$, the 1D complex (or absorption-mode) $\vec{p}$-projections of the spectrum, Eqs. [8] and [9], can now be expressed via the matrix element of $\hat{G}_{\vec{p}}(F)$:

$$I_{\vec{p}}(F) = (\Phi(0)|\hat{G}_{\vec{p}}(F)\Phi(0)), \qquad [49]$$

$$A_{\vec{p}}(F) = \mathrm{Im}\{I_{\vec{p}}(F)\}. \qquad [50]$$

Given matrix representations $\mathbf{U}_l$ for $l = 0, 1, 2, \ldots, \mathrm{D}$ in a Fourier window basis $\Psi(\vec{\varphi})$, Eqs. [40]–[42], numerical implementation of the Green's function approach is based on the following steps:

(i) Solve independently D generalized eigenvalue problems for $l = 1, 2, \ldots, \mathrm{D}$,

$$\mathbf{U}_l\mathbf{B}_{lk} = u_{lk}\mathbf{U}_0\mathbf{B}_{lk},$$
$$\mathbf{B}_{lk}^{\mathrm{T}}\mathbf{U}_0\mathbf{B}_{lk} = 1, \qquad [51]$$

to obtain the eigenvalues $u_{lk} \equiv e^{-i\tau_l\omega_{lk}}$ and eigenvectors $\mathbf{B}_{lk}$.

(ii) Use ω_{lk} and $\mathbf{B}_{lk}$ to construct a matrix representation of $\hat{\Omega}(\vec{p})$ in the basis of $\Psi(\vec{\varphi})$,

$$\mathbf{\Omega}_{\vec{p}} = \frac{1}{\tau_1}\sum_l p_l\tau_l\mathbf{\Omega}_l$$
$$= \frac{1}{\tau_1}\sum_l p_l\tau_l \sum_k \omega_{lk}\mathbf{U}_0\mathbf{B}_{lk}\mathbf{B}_{lk}^{\mathrm{T}}\mathbf{U}_0,$$

where $\mathbf{\Omega}_l$ are the corresponding matrix representations of $\hat{\Omega}_l$ in the Fourier window basis.

(iii) Solve another generalized eigenvalue problem

$$\mathbf{\Omega}_{\vec{p}}\mathbf{B}_{\vec{p}k} = \omega_{\vec{p}k}\mathbf{U}_0\mathbf{B}_{\vec{p}k},$$
$$\mathbf{B}_{\vec{p}k}^{\mathrm{T}}\mathbf{U}_0\mathbf{B}_{\vec{p}k} = 1. \qquad [52]$$

(iv) The frequencies $\omega_{\vec{p}k}$ and the amplitudes $d_{\vec{p}k}$ (see Eq. [43]) are then used in either Eq. [8] or Eq. [9] (with d_k replaced by $d_{\vec{p}k}$) to compute accordingly $I_{\vec{p}}(F)$ or $A_{\vec{p}}(F)$.

Note that we do not need to use Eq. [27] here as $\mathbf{U}_0$ appears in the right-hand side of the generalized eigenvalue problem (Eq. [52]), which requires only normalization of the eigenvectors $\mathbf{B}_{\vec{p}k}$.

The outlined procedure is numerically stable and fast. The fact that there may be many degenerate frequencies among $\omega_{\vec{p}k}$ corresponding, for instance, to the collapsed multiplets in the 45° projection of a 2D J spectrum, does not make either $I_{\vec{p}}(F)$ or $A_{\vec{p}}(F)$ nonunique. Generally, if there are several degenerate eigenvalues $\omega_{\vec{p}k}$ corresponding to a particular collapsed multiplet, the individual eigenvectors $\mathbf{B}_{\vec{p}k}$ are not well defined. However, the whole degenerate subspace of these eigenvectors, here called the *single-multiplet subspace*, is unique, subject to an orthogonal complex symmetric transformation within this subspace. This is sufficient for the correct spectral reconstruction using Eqs. [8] or [9] because the sum of amplitudes $\Sigma_k d_{\vec{p}k}$ over the single-multiplet subspace is invariant under any orthogonal transformation of the eigenvectors $\mathbf{B}_{\vec{p}k}$ within the subspace.

Since the eigenvectors within a degenerate subspace are not defined they cannot possibly be used in Eq. [45] to estimate the eigenvalues $u_{lk} = e^{-i\tau_l\omega_{lk}}$. However, as we demonstrated in Ref. (*10*) for the case of 2D J spectroscopy, one can uncover the multiplet frequencies along t_l (e.g., the acquisition time

dimension t_1) by solving the corresponding eigenvalue problem, Eq. [51], with the matrices $\mathbf{U}_l$ and $\mathbf{U}_0$ evaluated in the reduced basis of the single-multiplet subspace. The resulting eigenvalues $u_{lk} = e^{-i\tau_l\omega_{lk}}$ then all belong to this multiplet while the eigenvectors $\mathbf{B}_{lk}$ according to Eq. [43] define the individual amplitudes d_{lk}. These can be used to construct the multiplet cross sections along the lth dimension (*10*).

Reconstruction of a Multidimensional Spectrum Using a Multidimensional Green's Function

In this subsection we explore the possibility of constructing a multidimensional spectrum in the situation when the line list $\{\vec{\omega}_k, d_k\}$ is not unique or, at least, its generation is very difficult and therefore neither Eq. [6] or [7] can be used. As was already discussed, this ambiguity of the line list may be caused by either the existence of degenerate frequencies or, more generally, by noise. We will show that in the frame of FDM a well-behaved D-dimensional ersatz spectrum can be generated very efficiently from eigenvalues $u_{lk} \equiv e^{-i\tau_l\omega_{lk}}$ and eigenvectors $\mathbf{B}_{lk}$ computed by solving Eq. [51]. Here again we do not require the D sets of eigenvectors to be the same, a fact which is indicated by the additional subscript l; neither do we make an attempt to construct a unique, albeit approximate, set of eigenvectors by solving the D eigenvalue systems simultaneously (*9*).

An lth eigenvalue system yields its line list, i.e., the frequencies ω_{lk} and the amplitudes d_{lk}. The latter are obtained using Eqs. [43] modified by adding the subscript l, i.e.,

$$\sqrt{d_{lk}} = \sum_{\vec{\varphi}} [\mathbf{B}_{lk}]_{\vec{\varphi}} \sum_{n_1=0}^{M_1} \sum_{n_2=0}^{M_2} \dots \sum_{n_D=0}^{M_D} e^{i\vec{n}\vec{\varphi}} c(\vec{n}), \qquad [53]$$

where $\mathbf{B}_{lk}$ satisfies the relation

$$\Upsilon_{lk} = \sum_{\vec{\varphi}} [\mathbf{B}_{lk}]_{\vec{\varphi}} \Psi(\vec{\varphi}) \qquad [54]$$

between the eigenvectors Υ_{lk} of $\hat{U}_l$ and Fourier basis functions $\Psi(\vec{\varphi})$.

Generalizing Eq. [49] we can now write an example of a 2D complex spectral projection in terms of a 2D Green's function (*11*),

$$\begin{aligned} I(F_1, F_2) &= (\Phi(0)|\hat{G}_1(F_1)\hat{G}_2(F_2)\Phi(0)) \\ &\equiv \left(\Phi(0) \left| \frac{1}{\hat{\Omega}_1 - 2\pi F_1} \frac{1}{\hat{\Omega}_2 - 2\pi F_2} \Phi(0) \right. \right) \\ &= \sum_{k,k'} \frac{D_{kk'}}{(\omega_{1k} - 2\pi F_1)(\omega_{2k'} - 2\pi F_2)}, \end{aligned} \qquad [55]$$

with the cross-amplitudes $D_{kk'}$ defined by

$$D_{kk'} = \mathbf{B}_{1k}^{\mathrm{T}} \mathbf{U}_0 \mathbf{B}_{2k'} \sqrt{d_{1k} d_{2k'}}. \qquad [56]$$

This follows from the relation

$$(\Upsilon_{1k}|\Upsilon_{2k'}) = \mathbf{B}_{1k}^{\mathrm{T}} \mathbf{U}_0 \mathbf{B}_{2k'}, \qquad [57]$$

which is, in turn, a consequence of Eq. [54]. For a noiseless signal with no degeneracies the two sets of eigenvectors are exactly the same and the cross-amplitude matrix $D_{kk'}$ can be rearranged into a diagonal form. In such a case Eq. [55] boils down to Eq. [6]. In general, however, it is not diagonal due to degeneracies or noise or both. It is easy to see that in the case of a perfect noiseless signal, when a simultaneous diagonalization and computation of a unique set of eigenvectors is, in principle, possible, Eq. [55] takes care of the degeneracies, as it is invariant under orthogonal transformations within degenerate subspaces of either of the two Hamiltonians $\hat{\Omega}_1$, $\hat{\Omega}_2$. This means that for construction of a 2D spectrum we can avoid the conceptually nontrivial and computationally time-consuming simultaneous diagonalization.

To this end we give an example of a 2D double-absorption spectral projection modifying Eq. [55] accordingly,

$$\begin{aligned} A(F_1, F_2) = \sum_{k,k'} \mathrm{Re}\{D_{kk'}\} \mathrm{Im}\left\{ \frac{1}{\omega_{1k} - 2\pi F_1} \right\} \\ \times \mathrm{Im}\left\{ \frac{1}{\omega_{2k'} - 2\pi F_2} \right\}. \end{aligned} \qquad [58]$$

Note also another possible double-absorption spectral representation,

$$A(F_1, F_2) = \sum_{k,k'} \mathrm{Im}\left\{ \frac{D_{kk'}}{\omega_{1k} - 2\pi F_1} \right\} \mathrm{Im}\left\{ \frac{1}{\omega_{2k'} - 2\pi F_2} \right\}, \qquad [59]$$

which coincides with Eq. [58] in the case of all real cross-amplitudes $D_{kk'}$.

It is worth noting that Eqs. [55], [58], and [59] have, at least formally, direct product patterns while their derivation used the assumption of Eq. [2] with a nondirect product pattern. However, numerically most of the cross-amplitudes $D_{kk'}$ are nearly zero due to mutual orthogonality of most eigenvector pairs, so that the two representations are nearly equivalent.

Equations [55], [58], and [59] constitute a very important result as they allow the construction of unambiguous multidimensional spectra. An example of implementing Eq. [59] to a 2D J spectrum has been given in Ref. (*11*) and 2D HSQC spectrum in Paper II.

Phase Correction of the Multidimensional Spectrum

As was shown in Ref. (*7*) for the 1D case, FDM provides a simple means to correct phase distortion due to an initial time delay (*dead time*). For a multidimensional signal this is also straightforward. As such, for the 2D case, Eq. [59] is modified as

$$A(F_1, F_2) = \sum_{k,k'} \mathrm{Im}\left\{\frac{D_{kk'} e^{\,it_{d1}\omega_{1k} + it_{d2}\omega_{2k'}}}{\omega_{1k} - 2\pi F_1}\right\} \times \mathrm{Im}\left\{\frac{1}{\omega_{2k'} - 2\pi F_2}\right\}, \quad [60]$$

for time delays t_{d1} and t_{d2} with zero time corresponding to the first available signal data point. Equation [60] can be derived by noticing that only the cross-amplitudes $D_{kk'}$ should be corrected using

$$D_{kk'} \rightarrow D_{kk'} e^{\,it_{d1}\omega_{1k} + it_{d2}\omega_{2k'}}. \quad [61]$$

(3-1)D Simplification of 2D Spectra

Because multidimensional FDM works with the entire $N_{\mathrm{total}} = N_1 \times N_2 \times \cdots \times N_{\mathrm{D}}$ data set, useful projections can be obtained in which a very short additional time dimension is used to simplify a lower dimensionality spectrum. We have shown how a 1D proton-decoupled proton spectrum can be obtained from a 2D *J* experiment using only four time points in the *J* dimension (*10*) and then directly calculating the 45° projection. This principle is quite general and very useful. For example, as shown in Paper II, a proton–carbon HSQC spectrum can be condensed to a *singlet*-HSQC spectrum, in which each CH pair gives rise to a sharp singlet, by a 45° projection of a 3D HSQC *J* spectrum. This particular experiment makes no sense in the context of FT processing because (i) a large number of increments (e.g., 32–64) in the *J* dimension would be required to achieve the required resolution, leading to an unacceptably long experiment time; and (ii) the phase-sensitive 45° projection vanishes while the absolute-value projection shows poor resolution and lines with very wide wings. As such, each "singlet" in the FT spectrum can give contours as wide as the original multiplet that gave rise to it.

By contrast, a useful *singlet*-HSQC spectrum can be obtained using only *two* time points in the *J* dimension when FDM is employed. Very narrow absorption-mode resonances are obtained, increasing the resolution substantially. As it is unnecessary to record both N- and P-type spectra, the *singlet*-HSQC spectrum can in fact be obtained in the same total time as a conventional phase-sensitive HSQC spectrum (using FT processing).

Let us introduce the following convention for the 3D HSQC *J* spectrum, in which the $F_1 \times F_2$ plane corresponds to the HSQC spectrum, while the proton multiplets are rotated by 45° in the $F_1 \times F_3$ plane. In particular, the 2D part of the whole 3D signal $c(n_1\tau_1, n_2\tau_2, 0)$, roughly, corresponds to the conventional 2D HSQC signal, while $c(n_1\tau_1, 0, n_3\tau_3)$, the conventional 2D *J* signal of the protons directly bound to carbon-13.

A 2D complex *singlet*-HSQC spectral projection, where the proton multiplets of the conventional HSQC spectrum are collapsed into singlets at the proton chemical shifts, is here formally defined as the following 2D projection of the 3D spectrum,

$$I(F_{\vec{p}}, F_2) = \sum_k \frac{d_k}{(\omega_{1k} - \omega_{3k} - 2\pi F_{\vec{p}})(\omega_{2k} - 2\pi F_2)}. \quad [62]$$

Since implementation of this formula requires construction of a 3D line list, we rewrite it using Green's functions similar to those used above,

$$I(F_{\vec{p}}, F_2) = (\Phi(0)|\hat{G}_{\vec{p}}(F_{\vec{p}})\hat{G}_2(F_2)\Phi(0)) = \left(\Phi(0)\left|\frac{1}{\hat{\Omega}_{\vec{p}} - 2\pi F_{\vec{p}}}\frac{1}{\hat{\Omega}_2 - 2\pi F_2}\Phi(0)\right.\right), \quad [63]$$

where the first Green's function corresponds to the 45° projection in the 2D *J* plane with $\vec{p} = (\tau_1, 0, -\tau_1)$ and $\hat{\Omega}_{\vec{p}} = \hat{\Omega}_1 - \hat{\Omega}_3$, while the second projects to the carbon-13 chemical shift dimension.

Given matrix representations $\mathbf{U}_l$ for $l = 0, 1, 2, 3$ in a Fourier window basis $\Psi(\vec{\varphi})$ (Eqs. [40]–[42]), implementation of Eq. [63] is summarized by the following steps.

(i) Solve independently the three generalized eigenvalue problems with $l = 1, 2, 3$,

$$\mathbf{U}_l\mathbf{B}_{lk} = u_{lk}\mathbf{U}_0\mathbf{B}_{lk}, \qquad \mathbf{B}_{lk}^{\mathrm{T}}\mathbf{U}_0\mathbf{B}_{lk} = 1, \quad [64]$$

to obtain the eigenvalues $u_{lk} \equiv e^{\,-i\tau_l\omega_{lk}}$ and eigenvectors $\mathbf{B}_{lk}$.

(ii) Use $\{\omega_{1k}, \mathbf{B}_{1k}\}$ and $\{\omega_{3k}, \mathbf{B}_{3k}\}$ to construct a matrix representation of $\hat{\Omega}_{\vec{p}} = \hat{\Omega}_1 - \hat{\Omega}_3$ in the basis of $\Psi(\vec{\varphi})$,

$$\mathbf{\Omega}_{\vec{p}} = \mathbf{\Omega}_1 - \mathbf{\Omega}_3 = \mathbf{U}_0 \sum_k (\omega_{1k}\mathbf{B}_{1k}\mathbf{B}_{1k}^{\mathrm{T}} - \omega_{3k}\mathbf{B}_{3k}\mathbf{B}_{3k}^{\mathrm{T}})\mathbf{U}_0.$$

(iii) Solve another generalized eigenvalue problem,

$$\mathbf{\Omega}_{\vec{p}}\mathbf{B}_{\vec{p}k} = \omega_{\vec{p}k}\mathbf{U}_0\mathbf{B}_{\vec{p}k}, \qquad \mathbf{B}_{\vec{p}k}^{\mathrm{T}}\mathbf{U}_0\mathbf{B}_{\vec{p}k} = 1. \quad [65]$$

(iv) To evaluate the complex $I(F_{\vec{p}}, F_2)$ and double-absorption $A(F_{\vec{p}}, F_2)$ *singlet*-HSQC spectra, use the formulas

$$I(F_{\vec{p}}, F_2) = \sum_{k,k'} \frac{D_{kk'}}{(\omega_{\vec{p}k} - 2\pi F_{\vec{p}})(\omega_{2k'} - 2\pi F_2)}, \quad [66]$$

$$A(F_{\vec{p}}, F_2) = \sum_{k,k'} \mathrm{Im}\left\{\frac{D_{kk'}}{\omega_{\vec{p}k} - 2\pi F_{\vec{p}}}\right\} \times \mathrm{Im}\left\{\frac{1}{\omega_{2k'} - 2\pi F_2}\right\}, \quad [67]$$

where the cross-amplitudes $D_{kk'}$ are

$$D_{kk'} = \sqrt{d_{\vec{p}k}d_{2k}}\mathbf{B}^{\mathrm{T}}_{\vec{p}k}\mathbf{U}_0\mathbf{B}_{2k'},$$

$$\sqrt{d_{\vec{p}k}} = \sum_{\vec{\varphi}} [\mathbf{B}_{\vec{p}k}]_{\vec{\varphi}} \sum_{n_1=0}^{M_1}\sum_{n_2=0}^{M_2}\sum_{n_3=0}^{M_3} e^{i\vec{n}\vec{\varphi}}c(\vec{n}),$$

$$\sqrt{d_{2k}} = \sum_{\vec{\varphi}} [\mathbf{B}_{2k}]_{\vec{\varphi}} \sum_{n_1=0}^{M_1}\sum_{n_2=0}^{M_2}\sum_{n_3=0}^{M_3} e^{i\vec{n}\vec{\varphi}}c(\vec{n}).$$

Analogous formulas apply to a double projection of a 4D signal resulting in a simplified 2D spectrum. For example, as shown in Paper II a TOCSY spectrum, in which all 2D multiplets are completely decoupled, collapsing to singlets, can be obtained by recording additional two data sets with a single J increment in each dimension and then using a double 45° projection.

REDUCTION OF NOISE ARTIFACTS BY FILTER DIAGONALIZATION AVERAGING

An application of multidimensional FDM to noisy data is never free from imperfections, such as spurious spikes, or not well converged amplitudes d_k and their phases. In addition, Eq. [59], for example, should formally lead to a direct product pattern with possible peaks at all points $(\omega_{1k}, \omega_{2k'})$ with k, $k' = 1, 2, \ldots, K_{\mathrm{win}}$ within each processed frequency window. Although the corresponding amplitudes $D_{kk'}$ must ideally be zero for most cross peaks, they are not in practice, i.e., the ersatz spectrum constructed using Eq. [59] does contain many artifacts, which could, in principle, be big depending on how well the model of Eq. [2] fits the data. It might seem, by obvious analogy with various implementations of LP, that one could improve the appearance of the FDM ersatz spectrum by analyzing each spectral feature and deciding according to some criterion whether it should be accepted or discarded. Unfortunately, our experience is quite negative in this regard, as any such method seems to work only with high SNR and in the case of narrow and isolated resonances. In this case the extra peaks are completely unobtrusive in any event. Hence, we give up the idea of throwing away poles.

However, another much less aggressive and seemingly simple procedure to improve the appearance of the ersatz spectra exists (*7, 11*). It is based on multiple applications of FDM to nested subsets of the same signal, using a progressively larger total number of time points in successive calculations. The idea here is that the true signal poles are more stable with respect to the fitting parameters than the noise poles and artifacts. In particular, the phases of the unwanted features are sensitive to any change of the signal size, leading to their random appearance in the ersatz spectra. As such they can be "averaged out" by summing several ersatz spectra computed from the signals of various sizes. Probably other FDM parameters could be used in the same context, if their variation affected the appearance of the noise and artifacts, although we found that varying signal size is a robust procedure. When multidimensional NMR signals are processed with FDM, a natural parameter for averaging is the maximum processing time along the acquisition time dimension n_1.

Note, though, another more elegant procedure, the *pseudo-noise averaging*, discovered after this paper was submitted for publication. It is based on exploiting the great sensitivity of the output (ersatz spectrum) to the small variations in the input (time signal). Namely, perturbation of the signal by a small amount $\delta(\vec{n})$ leads to large fluctuations, $\Delta(\vec{F})$, in the output FDM spectrum. This is schematically shown by the diagram

$$\boxed{c(\vec{n}) + \delta(\vec{n})} \rightarrow \boxed{\mathrm{FDM}} \rightarrow \boxed{I_\delta(\vec{F}) = I_0(\vec{F}) + \Delta(\vec{F})}. \quad [68]$$

The fluctuations $\Delta(\vec{F})$ have essentially random appearance, so the artifacts are removed by averaging $I_\delta(\vec{F})$ over sufficiently many realizations of the pseudo-noise $\delta(\vec{n})$. It turns out that there is a great flexibility in choosing the distribution and the amplitude of the pseudo-noise $\delta(\vec{n})$ (as long as $\langle\delta(\vec{n})\rangle = 0$ and $\langle|\delta(\vec{n})|^2\rangle$ is sufficiently large) to give unique and artifact-free averaged spectrum.

For both types of the FDM averaging described above the bad news is that the computational effort is now multiplied by N_{FDM}, the number of the single FDM calculations, while sometimes leading to only moderate improvements (a factor of $\sqrt{N_{\mathrm{FDM}}}$ reduction of the artifacts, assuming their random appearance). The good news is that a single FDM calculation is reasonably fast so that we can still afford such an increase of the CPU time, even if not expecting a huge improvement. Thus FDM averaging should be customary in all multidimensional FDM calculations unless one finds a less time-consuming solution in the future.

SUMMARY

The present paper describes several important developments of the multidimensional FDM. Expressions for construction of absorption-mode spectra from purely phase-modulated NMR signals based on pure linear algebra are derived. In particular, FDM allows direct calculation of various reduced dimensionality spectral projections leading under certain conditions to significant simplifications of the spectra and avoiding calculation of both the whole multidimensional spectrum and the complete multidimensional line list. The latter seemed to be a prerequisite in the earlier works on 2D FDM (*4–6, 9*).

Even though the multidimensional line list, i.e., the set $\{\vec{\omega}_k, d_k\}$ with $\vec{\omega}_k = (\omega_{1k}, \omega_{2k}, \ldots, \omega_{\mathrm{D}k})$, is highly desired, its construction for severely truncated and noisy signals may be complicated by some conceptual problems originated from the ill-defined nature of the HIP, Eq. [2]. In the FDM framework the problem of constructing the multidimensional line list arises when the computed eigenbases $\{Y_{lk}\}$ of the evolution operators $\hat{U}_l = e^{-i\tau_l\hat{\Omega}_l}$ (where the l labels the time or frequency dimension) appear to be incompatible with each other, preventing construction of a unique basis set $\{Y_k\}$. An lth basis set with the corresponding eigenfrequencies ω_{lk} can unambigu-

ously define only the lth projection $\{\omega_{lk}, d_{lk}\}$ of the line list, while the problem of coupling the different projected frequencies ω_{lk} into a single vector $\vec{\omega}_k$ remains unsolved for the general case of truncated and noisy data.

The new Green's function approach to construct the FDM spectra, e.g., based on using Eqs. [59] and [67], overcomes the problem of ambiguity of the line list. However, the resulting spectra still have some artifacts due to the said ill-defined nature of the problem. Because these artifacts are very sensitive to either parameters of the FDM calculations or small variations in the input data, averaging allows us to reduce them below the acceptable level. Unfortunately, the FDM averaging is quite expensive and therefore might not be the optimal solution.

To conclude, in the future we will revisit these issues, namely, the line list construction and the problem of avoiding numerically expensive FDM averaging.

APPENDIX

Some Important Notations and Definitions

$\vec{n} \equiv (n_1\tau_1, n_2\tau_2, \ldots, n_D\tau_D)$	D-dimensional time vector
$c(\vec{n}) \equiv c(n_1\tau_1, n_2\tau_2, \ldots, n_D\tau_D)$	NMR signal defined on an evenly spaced D-dimensional time grid
$c(\vec{n}) = \sum_k d_k e^{-i\vec{n}\vec{\omega}_k} \equiv \sum_k d_k \exp[-i\sum_{l=1}^{D} n_l\tau_l\omega_{lk}]$	D-dimensional harmonic inversion problem
$\vec{\omega}_k \equiv (\omega_{1k}, \omega_{2k}, \ldots, \omega_{Dk})$, $\omega_{lk} = 2\pi f_{lk} - i\gamma_{lk}$	Vector of complex frequencies
$\{\vec{\omega}_k, d_k\}$	Line list of vector frequencies and amplitudes
$I(\vec{F}) \equiv I(F_1, F_2, \ldots, F_D) = \sum_k d_k \prod_{l=1}^{D} \frac{1}{\omega_{lk} - 2\pi F_l}$	A D-dimensional complex ersatz spectrum
$I_{\vec{p}}(F) = \sum_k \frac{d_k}{\vec{p}\vec{\omega}_k/\tau_{\max} - 2\pi F} \equiv \sum_k \frac{d_k}{\omega_{\vec{p}k} - 2\pi F}$	1D spectral projection along vector $\vec{p}$
$(\Psi\|\Phi) = (\Phi\|\Psi)$	Complex symmetric inner product
$\hat{\Omega}_l = \sum_k \omega_{lk}\|\Upsilon_k)(\Upsilon_k\|$	Commuting complex symmetric Hamiltonian operators
ω_{lk} and Υ_k	Eigenvalues and eigenvectors of $\hat{\Omega}_l$
$\vec{\Omega} \equiv (\hat{\Omega}_1, \hat{\Omega}_2, \ldots, \hat{\Omega}_D)$	Operator vector
$\hat{U}(\vec{n}) \equiv e^{-i\vec{n}\vec{\Omega}} \equiv \exp[-i\sum_{l=1}^{D} n_l\tau_l\hat{\Omega}_l]$	Discrete time evolution operator
$u_k(\vec{n}) \equiv e^{-i\vec{n}\vec{\omega}_k}$ and Υ_k	Eigenvalues and eigenvectors of $\hat{U}(\vec{n})$
$\Phi(0) = \sum_k \sqrt{d_k}\Upsilon_k$	Initial state
$c(\vec{n}) = (\Phi(0)\|\hat{U}(\vec{n})\Phi(0))$	Quantum mechanical ansatz
$\Phi(\vec{n}) = \hat{U}(\vec{n})\Phi(0)$, $n_l = 0, 1, \ldots, M_l$	Subspace of Krylov vectors
$\Upsilon_k = \sum_{\vec{n}} [\mathbf{B}_k]_{\vec{n}}\Phi(\vec{n})$	Expansion of the eigenvectors in terms of the Krylov vectors
$[\mathbf{U}(\vec{p})]_{\vec{n}\vec{m}} \equiv (\Phi(\vec{n})\|\hat{U}(\vec{p})\Phi(\vec{m})) = c(\vec{n} + \vec{m} + \vec{p})$	Matrix elements of the evolution operator in the primitive Krylov basis
$[\mathbf{U}_0]_{\vec{n}\vec{m}} \equiv (\Phi(\vec{n})\|\Phi(\vec{m})) = c(\vec{n} + \vec{m})$	Overlap matrix elements
$\mathbf{U}(\vec{p} + \vec{q})\mathbf{B}_k = u_k(\vec{p})\mathbf{U}(\vec{q})\mathbf{B}_k$, $\mathbf{B}_k^{\mathrm{T}}\mathbf{U}_0\mathbf{B}_{k'} = 1$	Generalized eigenvalue problem with the normalization condition
$\sqrt{d_k} = (\Upsilon_k\|\Phi(0)) = \sum_{\vec{n}} [\mathbf{B}_k]_{\vec{n}} c(\vec{n})$	The amplitudes in terms of the eigenvectors
$\Psi(\vec{\varphi}) = \sum_{n_1=0}^{M_1}\sum_{n_2=0}^{M_2}\ldots\sum_{n_D=0}^{M_D} e^{i\vec{n}\vec{\varphi}}\Phi(\vec{n})$	Fourier basis function defined at $\vec{\varphi} \equiv (\varphi_1, \varphi_2, \ldots, \varphi_D)$
$\Upsilon_k = \sum_{\vec{\varphi}} [\mathbf{B}_k]_{\vec{\varphi}}\Phi(\vec{\varphi})$	Expansion of the eigenvectors in the Fourier basis
$[\mathbf{U}(\vec{p})]_{\vec{\varphi}\vec{\varphi}'} \equiv (\Psi(\vec{\varphi})\|\hat{U}(\vec{p})\Psi(\vec{\varphi}'))$	Matrix elements of the evolution operator in the Fourier basis
$\hat{\Omega}_{\vec{p}} = \frac{1}{\tau_1}\vec{p}\vec{\Omega} = \sum_k \omega_{\vec{p}k}\|\Upsilon_{\vec{p}k})(\Upsilon_{\vec{p}k}\|$	$\vec{p}$-projection of the Hamiltonian vector and its spectral representation

$$\hat{G}_{\vec{p}}(F) \equiv \frac{1}{\hat{\Omega}_{\vec{p}} - 2\pi F} = \sum_k \frac{|\Upsilon_{\vec{p}k})(\Upsilon_{\vec{p}k}|}{\omega_{\vec{p}k} - 2\pi F}$$ $\vec{p}$-projection of the Green's function and its spectral representation

$$I_{\vec{p}}(F) = (\Phi(0)|\hat{G}_{\vec{p}}(F)\Phi(0))$$ 1D complex spectral projection along $\vec{p}$ using the Green's function

$$I(F_1, F_2) = (\Phi(0)|\hat{G}_1(F_1)\hat{G}_2(F_2)\Phi(0))$$ A 2D complex spectral projection using the 2D Green's function

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