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QUANTUM-MECHANICAL SIGNAL PROCESSING AND SPECTRAL ANALYSIS

DŽEVAD BELKIĆ

ROUTLEDGE

Quantum-Mechanical Signal Processing and Spectral Analysis

About the author

Dževad Belkić is a theoretical physicist. He is Professor of Mathematical Radiation Physics at Karolinska Institute in Stockholm, Sweden. His current research activities are in atomic collision physics, radiation physics, radiobiology, magnetic resonance physics and mathematical physics. In atomic collision physics, he has worked on many problems including major challenges such as the theory of charge exchange and ionization at high non-relativistic energies. *Inter alia* he used distorted wave methods, paying special attention to treatments with correct boundary conditions for scattering particles which interact through Coulomb potentials.

In radiation physics, Professor Belkić has worked on the passage of fast electrons and multiply charged ions through tissue as needed in radiation therapy in medicine. Here he has employed both deterministic methods through the Boltzmann equation and stochastic simulations via Monte Carlo computations. In radiobiology, he has worked on mathematical modelling for cell survival, and has focused on mechanistic modelling by including the main pathways for survival of cells under irradiation during radiotherapy.

In magnetic resonance physics, Professor Belkić has worked on nuclear magnetic resonance in medicine where he focused on high-resolution parametric signal processors which go beyond the conventional shape estimations of spectra. In mathematical physics, he has worked on many problems including the derivation of analytical expressions for scattering integrals or bound-free form factors, for rational response functions in signal processing, for coupling parameters in the nearest neighbour approximation which is one of the most frequently used methods in physics and chemistry, etc.

He has published more than 160 scientific publications which have received over 2000 citations. His book *Principles of Quantum Scattering Theory* has been published in 2004 by the Institute of Physics Publishing. He has received a number of international awards including the triple Nobel grantee status for research grants in atomic collision theory from the Royal Swedish Academy of Sciences as approved by the Nobel Committee for Physics.

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Preface

This book is on quantum-mechanical signal processing based upon the Padé approximant (PA). We here link the PA and the Lanczos algorithm to design the Padé–Lanczos approximant (PLA). The PLA is operationalized with the recursive algorithm called the fast Padé transform (FPT) for both parametric and non-parametric estimations of spectra. The FPT for any given power series is defined by the unique quotient of two polynomials. This processor provides a meaningful result even when the original expansion diverges. It can significantly accelerate slowly converging sequences/series. As opposed to a single polynomial, e.g. the fast Fourier transform (FFT), the FPT can analytically continue general functions outside their definition domains. Moreover, we show that the FPT is an efficient solver of generalized eigenproblems, e.g. the quantum-mechanical evolution/relaxation matrix $\mathbf{U}$ comprised of auto-correlation functions. These generic functions can be either computed theoretically or measured experimentally. Such a concept, put forward as a computational tool, surpasses its initial purpose. Indeed auto-correlation functions represent a veritable alternative formulation of quantum mechanics. This is not just because all the major observables, e.g. complete energy spectra, local density of states, quantal rate constants, etc, are expressible through the auto-correlation functions. It is also because these and other observables could be given completely in terms of some appropriate, relatively small informational parts that can be singled out and analysed separately from the unwanted/redundant remainder of the full data set of auto-correlation functions. The required dimensionality reduction of original large problems treated by the FPT can be achieved by, e.g. windowing using the band-limited decimation. Alternatively, as done in this book, the Lanczos tridiagonalization can be employed yielding sparse Jacobi matrices in terms of the Lanczos coupling parameters $\{\alpha_n, \beta_n\}$ that have very important physical interpretations. The FPT is naturally ingrained in the Schrödinger picture of quantum mechanics and in the total time-independent Green function for the studied system. This yields a versatile framework for a unified treatment of spectroscopy and collisions within signal processing and quantum mechanics. In the quantum-mechanical method of nearest neighbours or tight bindings, we use time signal points $\{c_n\}$ as the only input data to derive the exact analytical expressions for the FPT, the continued fractions, the Lanczos polynomials

$\{P_n(\omega), Q_n(\omega)\}$, the couplings $\{\alpha_n, \beta_n\}$, the Lanczos state vectors ψ_n, the total wavefunction $\Upsilon(\omega)$ at any frequency ω and the 'Hamiltonian' operator $\hat{\Omega}$.

Within signal processing we also analyse ordinary difference equations, base-quotient concept, iterative relaxations, nonlinear accelerations, rational approximations, regularizations of spurious roots and the methods of moments. The analysis is performed within the context of the so-called harmonic inversion problem, or equivalently, the spectral decomposition or quantification problem. This is an inverse/reconstruction problem which is aimed at retrieving uniquely the whole information from the input time signal $\{c_n\}$, such that the output data contain the number K, position $\mathrm{Re}(\omega_k)$, height $|d_k|$, width $\mathrm{Im}(\omega_k)$ and phase $\mathrm{Arg}(d_k)$ of every complex harmonic including all possible degeneracies/multiplicities of normal mode frequencies $\{\omega_k\}$ due to overlapping resonances[1]. The FPT has the unique virtues of unequivocally providing all these parameters for the non-degenerate and degenerate spectra with the least computational effort by attaining the required accuracy via robust and stable signal processing. This could be fully accomplished in only two simple steps: (1) solving a system of linear equations to extract the expansion/prediction coefficients of the characteristic/secular polynomial $Q_K(\omega)$ directly from $\{c_n\}$ and (2) rooting $Q_K(\omega)$ to obtain all the sought complex frequencies $\{\omega_k\}$ $(1 \leq k \leq K)$. Only step (1) is needed in the non-parametric FPT, which is given by the polynomial quotient $P_L(\omega)/Q_K(\omega)$. This yields the shape spectrum in the FPT at any ω. In the quantification problem, the parametric FPT is required by carrying out step (2) which gives all the K complex frequencies $\{\omega_k\}$. The corresponding complex amplitudes $\{d_k\}$ are obtained from the two separate explicit expressions for the non-degenerate and degenerate spectrum as the residues of $P_L(\omega)/Q_K(\omega)$ taken at one single frequency $\omega = \omega_k$. This produces the K isolated Lorentzian resonances in a non-degenerate spectrum if the roots $\{\omega_k\}$ of $Q_K(\omega)$ are all distinct (no multiplicities). When some of the roots $\{\omega_k\}$ of $Q_K(\omega)$ coincide, i.e. have multiplicities, a degenerate non-Lorentzian spectrum $P_L(\omega)/Q_K(\omega)$ is obtained in the FPT which describes all isolated and completely or partially overlapping resonances. By contrast, other parametric methods, e.g. linear predictor (LP) or the Hankel–Lanczos singular value decomposition (HLSVD), are much more computationally demanding as they obtain the set $\{d_k\}$ by solving another system of linear equations using *all* the elements $\{\omega_k\}$ $(1 \leq k \leq K)$, where errors in some $\{\omega_k\}$ might cause severe inaccuracies in d_k. Moreover, to arrive at d_k the method of filter diagonalization (FD) performs additional computations for the full state vectors $\{\Upsilon_k\}$ from the generalized eigenproblem of the evolution/relaxation matrix, i.e. the Hankel data matrix. Relative to the linear FFT, the advantages of the nonlinear FPT are in the significantly increased resolution for the same signal length or in the same resolution for shorter signal lengths. Furthermore, as opposed to other nonlinear

[1] Hereafter the symbols $\mathrm{Re}(z)$ and $\mathrm{Im}(z)$, respectively, denote the real and imaginary parts of a complex number z.

methods that typically undergo wild oscillations before they eventually stabilize, the FPT exhibits a strikingly stable convergence with the increased signal length. This is the present finding via an illustrative spectral analysis of a time signal, which has been experimentally measured via Magnetic Resonance Spectroscopy (MRS) at the magnetic field strength of 7T encoded from the brain of a healthy volunteer.

We also analyse unphysical (spurious or extraneous) resonances in the parametric estimation of spectra. Spurious peaks stem from noise corruption of the time signal and from the so-called overdetermination problem. If the signal of length N happens to have less than $N/2$ peaks, the problem becomes algebraically overdetermined, since there are more equations than unknowns. This leads to singular values associated with false peaks that represent spurious resonances. In the PA, extraneous roots could appear in both the numerator and denominator polynomial. Spurious roots in the denominator polynomial of the PA are undesirable, since they lead to unphysical spikes in the Padé spectrum. Spurious roots in the numerator polynomial of the PA are also unwelcome, since they fill in the valleys with unphysical anti-resonances and this destroys the phase minimum as well as the uniqueness feature of the PA. We examine this important problem by using the so-called constrained root reflection, which is an analytical procedure for regularizing spurious roots. First, we separate unequivocally the genuine from spurious resonances in the Padé power spectrum, which is itself the Padé–Chebyshev approximant (PCA). Second, the unstable PCA containing diverging and converging exponentials is properly stabilized. This is done by a special root reflection, which reverses the sign of diverging exponentials, so that they are relocated on the side of genuine resonances. Such a procedure is accomplished under the constraint that the parameter and the shape spectra of the PA are the same. The ensuing constrained root reflection has its physical justification in the preservation of the total energy of the signal via the Parseval identity. The resulting method is called the Padé–Schur approximant (PSA) which, as a stable estimator, possesses only converging exponentials. The PSA describes the Padé power spectrum as the unique ratio of two Schur polynomials whose computed roots are all adequately regularized by being placed on the side of physical resonances. Thus, rather than trying to eliminate or reduce the noise content from the measured data, as has often been attempted previously with a potential risk of losing weak genuine spectral features, the PSA processes noise, as measured, together with the physical signal.

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Acronyms used in the text

ABC:	Absorbing Boundary Conditions
AR:	Auto-Regressive
ARMA:	Auto-Regressive Moving Average
a.u.:	arbitrary units
bl:	band limited
bld:	band limited decimated
CF:	Continued Fractions
CCF:	Contracted Continued Fractions
CPU:	Central Processing Unit
CSI:	Chemical Shift Imaging
DFT:	Discrete Fourier Transform
DLP:	Decimated Linear Predictor
DOS:	Density of States
DPA:	Decimated Padé Approximant
DSD:	Decimated Signal Diagonalization
DVR:	Discrete Variable Representation
EHIP:	Extended Harmonic Inversion Problem
ESPRIT:	Estimation of Signal Parameters via Rotation Invariance
FD:	Filter Diagonalization
FFT:	Fast Fourier Transform
FID:	Free Induction Decay
FPT:	Fast Padé Transform
FWHM:	Full Width at the Half Maximum
GHIP:	Generalized Harmonic Inversion Problem
GSO:	Gram–Schmidt Orthogonalization
HIP:	Harmonic Inversion Problem
HLSVD:	Hankel–Lanczos Singular Value Decomposition
ICR:	Ion Cyclotron Resonance
IDFT:	Inverse Discrete Fourier Transform
IDOS:	Integrated density of states
LCF:	Lanczos Continued Fractions
LCModel:	Linear Combination of Model *in vitro* Spectra

lhs: left-hand side
LP: Linear Predictor
LPC: Linear Predictor Coding
MA: Moving Average
MR: Magnetic Resonance
MRI: Magnetic Resonance Imaging
MRS: Magnetic Resonance Spectroscopy
MRSI: Magnetic Resonance Spectroscopic Imaging
MUSIC: Multiple Signal Classification
NMR: Nuclear Magnetic Resonance
ODE: Ordinary Differential Equations
OΔE: Ordinary Difference Equations
OPA: Operator Padé Approximant
PA: Padé Approximant
PD: Product-Difference
PCA: Padé–Chebyshev Approximant
PLA: Padé–Lanczos Approximant
ppm: parts per million
PVL: Padé via Lanczos
PSA: Padé–Schur Approximant
RCM: Rotated Coordinate Method
RF: Radio Frequency
rhs: right-hand side
rms: root mean square
ROPEM: Recursive Orthogonal Polynomial Expansion Method
RRGM: Recursive Residue Generation Method
SPECT: Single Photon Emission Computerized Tomography
SVD: Singular Value Decomposition
TE: Echo Time
QD: Quotient-Difference

Chapter 1

Introduction

Among sciences and engineering marking the beginning of the new millennium, the versatile concept of *signals* and *images* ranks high along with a few outstanding leading strategies. This is because signals are indeed universal responses of objects to perturbations ranging from galactic interstellar phenomena, helio-seizmology to the central nervous system or to the genetic code, etc. Such responses can vary from, e.g. intercellular communications all the way down to subatomic particle levels and beyond. Besides having the unique potential to successfully intertwine and indeed unify some of the major interdisciplinary research fields in basic and applied sciences with the necessary bridging to engineering and technology, signals are also directly amenable to innumerable practical applications including the issue of the health of human beings. For an illustration, it suffices to mention only a few of the major fields where signals play a key role, e.g. molecular beam magnetic resonance [1], nuclear magnetic resonance (NMR) [2]–[6], ion cyclotron resonance (ICR) mass spectroscopy [7]–[9], magnetic resonance spectroscopy (MRS) [10]–[29], magnetic resonance imaging (MRI) [30]–[32], magnetic resonance spectroscopic imaging (MRSI) [33]–[39] which is also called chemical shift imaging (CSI), single photon emission computerized tomography (SPECT) [40]–[42], etc.

These unique tools for magnetic resonance (MR) physics are based upon inverse reconstruction problems that are deeply rooted in quantum-mechanical scattering and spectroscopy. Moreover, they represent a sequence of indispensable non-invasive diagnostic techniques of paramount importance in modern medical practice. This was made possible by a remarkable cooperation of mathematics, physics, medicine, computing science and technologies. Development of software was followed by hardware implementations and it is this coupling which represents the key driving force of signal and image processing. When it comes to imaging of, e.g. a patient, severe constraints must be put onto the applied magnetic field strengths that cause perturbations of the examined organ of the human body. These constraints might limit the resolution of the obtained spectra or images, and it is precisely here where recent vigorous advances in

the mathematical software become critical to further progress. However, all the commercial software built into, e.g. spectrometers is presently based upon the fast Fourier transform (FFT) [43]–[55], despite the availability of a myriad of high-resolution signal and image processing methods. The reason for this is that most of the existing parametric methods in signal processing are inherently *unstable* in the sense of Schur [56]. This means that some of the complex eigenvalues or roots of characteristic polynomials are unphysical, i.e. spurious within many parametric methods such as the linear predictor (LP) [57]–[64], the Padé approximant (PA) [65]–[92], the Lanczos algorithm [93]–[104], the multiple signal classification (MUSIC) [105], the estimation of signal parameters via rotation invariance technique (ESPRIT) [106], the Padé–Laplace transform [107], the filter diagonalization (FD) [108]–[112], the decimated signal diagonalization (DSD) [113]–[116], the fast Padé transform (FPT) [16]–[29], etc. In signal processing, these extraneous eigenvalues of, e.g. the evolution **U**-matrix lead to exponentially increasing harmonics of the corresponding free induction decay (FID) or time signal c_n with the increase of time. These diverging harmonics are also encountered in the decimated Padé approximant (DPA) and decimated linear predictor (DLP) [115] through the emergence of unstable, i.e. non-Schur polynomials with some of their roots located on the 'wrong side' of the unit circle with exploding exponentials as the time increases infinitely [56].

The main difficulty with spurious roots is not so much in that they are regularly encountered in practice, but rather it is the implementation of a theoretically devised procedure which, by construction, would guarantee that the spectral density (power or magnitude spectrum) is invariant under the selected regularization. Recently, the FD has been applied to a variety of problems [108]–[112], but it was found that this method also generates spurious frequencies with $\mathrm{Im}(\omega_k) > 0$ leading to exponentially diverging natural or fundamental harmonics in the time signal. However, both the FD and the DSD have a technique for handling spurious eigenenergies [108]–[116]. This procedure consists of comparing the eigenvalues of the powers of the matrix $\mathbf{U}^{(s)}$ (comprised of the matrix elements of the sth power of the evolution operator, $\hat{\mathrm{U}}^s$) for two values of the integer s, e.g. $s = 1$ and $s = 2$. From these two separate diagonalizations only those eigenvalues are retained that are within a prescribed level of accuracy and the other fluctuating solutions are rejected. Similarly, the recursive Lanczos algorithm [93]–[104] for solving large eigenproblems produces extraneous eigenvalues, but they are also managed by an adequate procedure [99]. In the Padé–Laplace transform [107] and DPA [115] spurious roots are recognized by comparing the results between the main diagonal and several successive paradiagonals of the Padé table. Within a prescribed threshold of accuracy, some of the roots are virtually unaltered when passing from the main diagonal to several neighbouring paradiagonals. These stable roots are classified as physical and the corresponding peaks are retained in the final spectrum. However, the same comparison also finds some peaks whose parameters change considerably beyond the prescribed accuracy level. These resonances are considered as unphysical, i.e.

spurious and they are discarded from the spectrum. Diagonalization of the matrix $\mathbf{U}^{(s)}$ in the DSD for an integer s is mathematically equivalent to the DPA for the main diagonal ($s = 1$) and the paradiagonals ($s > 1$). Therefore, the techniques for recognizing spurious resonances in the DSD or FD and DPA are the same as the procedure used previously in the Padé–Laplace transform [107].

As an alternative to the mentioned numerical techniques for dealing with extraneous resonances, we presently elaborate an *analytical* tool for unambiguous identifications of all existing spurious roots. To this end, within the generic PA, we use the so-called constrained root reflection [18] for regularization of *all* the encountered spurious roots. In particular, the procedure [18] automatically assures that *both* the numerator and denominator polynomials of the PA are stable or Schur polynomials [56]. This generalizes the related previous work from [117] where only the Padé denominator polynomial was regularized by the constrained root reflection. In [18], a variant of the root reflection has been analysed by subjecting all the spurious roots of the two Padé polynomials P_L/Q_K to complex conjugation, but with the important constraint that magnitude $|P_L/Q_K|$ and, likewise, power spectra $|P_L/Q_K|^2$ remain strictly unaltered. This is the constrained root reflection which regularizes all the encountered extraneous poles by relocating them to the site of the genuine poles. In other words, this method deliberately *avoids eliminating* any of the encountered spurious roots. This is because, in practice, some of the spurious roots might well be associated with the genuine, physical transients that were supposed to decay, but nevertheless remained in the signal for the given duration of the acquisition time of the FID. The presence of these 'spurious' resonances may influence the spectral information in the signal and, as such, they should not be eliminated from the analysis. The constrained root reflection as a power-spectrum-preserving regularization of spurious resonances should be contrasted to the usual root reflection, which replaces $\mathrm{Im}(\omega_k)$ by $-|\mathrm{Im}(\omega_k)|$ for $\mathrm{Im}(\omega_k) > 0$, but without a simultaneous requirement that the resulting magnitude and/or power spectra remain the same as if this replacement were absent.

Originally, signal and image processing developed autonomously, and remained for a long time mainly within the realm of applied sciences and engineering. Yet, here, very similar or nearly the same methods have been used as in the basic sciences. For example, in electric circuit theory [118]–[120], one of the most frequently used methods is the so-called rational response of the examined system to an external perturbation. This response is, in fact, the PA in the frequency domain. Precisely the same type of PA can also be found in various fields that use signal processing (speech patterns, system theory, optimizations, heart rate variability, etc) or in mathematical statistics under different names such as the auto-regressive moving average (ARMA) [121]–[129]. Reconstructing spectra and images from the received signals is a general inverse problem with its intrinsic time development, which is ideally suited for description by the Schrödinger picture of quantum mechanics. Therefore, it could be beneficial for signal and image processing to intertwine with a quantum-

mechanical description of stationary and time-independent phenomena. It has been only within a few recent years [108]–[116] that the situation improved through using quantum mechanics and quantum resonant scattering theory in signal processing, by relying upon the concept of auto-correlation functions as the amplitude probabilities for survival of the Schrödinger or Krylov states. In such an approach, the Schrödinger ordinary or generalized eigenvalue equations and the spectral problem of the resolvent or the Green operator play the pivotal roles. From this framework many more opportunities might open up with the advantage of furthering signal and image processing themselves, but also of providing new tools for studying generic spectra irrespective of their experimental or theoretical origins. The spin-offs from this cross fertilization of the invoked research fields is anticipated to have a twofold benefit, e.g. emergence of new high-resolution signal and image processors for applied sciences as well as engineering and, in turn, the usage of these very same methods in basic sciences for analysing spectra of large physical, chemical or biological systems. Naturally, the literature on signal and image processing in applied mathematics and engineering [118]–[123] is abundant with specially designed methods for robust performance in industry and technology under rigorous requirements for accuracy, stability and reliability. To achieve such strict goals the most advanced mathematical methods have been used with the possibility for export to applied and basic sciences for versatile applications. For this to happen in a manner which is systematic rather than sporadic, more cross disciplinary interactions are needed with the mutual benefits that would *inter alia* reduce unnecessary duplication of the results in non-overlapping fields. In addition to other existing efforts along these lines, the two recent topical issues on spectral analysis [130, 131] have already paved a new road for cross disciplinary exchange of ideas. The present book is also on spectral analysis with the main goal of attempting to convey to the reader some of the common aspects, strategies and mathematical methods in seemingly disjoint research fields.

Notwithstanding the importance of the practical implications in arriving at highly resolved spectra and images, the key merit of introducing quantum mechanics into signal processing is in providing the fundamental framework of a complete theory of physics. This is crucial due to the possibility of directly relating arbitrary signals to the *dynamics* of the examined system and its time evolution described by the first principles of physics. For overly complex systems the dynamics might be either unknown or unmanageable for direct theoretical treatments. In such cases, whenever experimental data are available as, e.g. counts per channel, time signals and the like, quantum mechanics can peer into the dynamics of the system and extract the full spectral information. This is possible due to, e.g. the established equivalence between time signals that lead to Lorentzian spectra and auto-correlation functions [108]–[115]. This equivalence replaces the usual nonlinear fitting problem of experimental data by the standard quantum-mechanical search for eigenspectra of the studied system. The obtained results reconstruct the unknown dynamics and interactions in the system which

has undergone certain transitions under the influence of a perturbation, before generating the recorded time signal. This is at once recognized as the well-studied inverse scattering problem in quantum mechanics, where one is given certain experimentally measured data (e.g. phase shift, etc) and the task is to retrieve the interaction potentials that are the key to every physics problem. It is this unfolded dynamics which provides the sought information about the studied system, e.g. the proton spin density $\rho(x, y)$ yielding the emission data in MRI, MRS, MRSI, etc. This accomplishes the task of locating the accurate spatial positions (x, y) of protons. It is a plot of the reconstructed observable $\rho(x, y)$ which directly gives an image of the object whose signals were acquired via, e.g. MRI or MRSI. Slices of such images can be spectroscopically analysed, as done within MRSI, to gain invaluable quantitative information about peak parameters, such as abundance of various metabolites, their relaxation times, etc. For example, relaxation times of lipids, whose peaks are superimposed on top of 'noisy' background, exhibit very different patterns in tumorous and healthy tissue, pointing at the important diagnostic role of MRS and MRSI in medicine [16]–[29], [33]–[39].

There is tremendous interest in medicine for MR physics whose multitude of retrieval or reconstruction techniques has nowadays attained the status of an essential part of diagnostic radiology. The ultimate goal of MRI is to generate two-dimensional images of pre-assigned sections of the examined human organ. The two distinct aspects of MRI are advantageous relative to other competitive methods: (i) an arbitrary orientation/position of the imaged area and (ii) a large contrast among soft tissues. In addition to imaging static anatomy, many current clinical tasks use MRI for imaging blood vessels without contrast agents, cardiac imaging, dynamic imaging of the musculoskeletal system as well as for measuring tissue temperature and recording diffusion in tissue. The common denominator in all the MR encoding techniques is radio frequency (RF) excitation of the sample and a reliance upon the elementary magnetic fields that originate from the nuclear magnetic moments. The Larmor precession of these magnetizations around the external constant magnetic field, combined with the additional magnetic field gradients and RF pulses, generates a small current in the receiver coil which surrounds the relevant part of the examined human body. This is the pathway through which the sample responds to the external perturbation, after which de-excitation of the sample takes place, leading to an echo with a composite electromagnetic time signal. Spectral and image analysis of this complex valued signal can reveal the types and the spatial two-dimensional positions (x, y) of the nuclei that are imaged within the selected sample.

The current key problems of MRI are long imaging times and insufficient spatial resolution. The reasons for such an unsatisfactory situation is that advances in MRI have thus far been tied to upgrading hardware, whereas the commercially implemented signal and image processing software is limited exclusively to the FFT. However, the FFT is known to be a low-resolution estimator unless exceedingly long time signals are recorded, which is in clinical practice precluded by high levels of noise and, more importantly, by intolerably long exposure of

patients to external magnetic fields. This situation could be significantly improved by simply complementing the FFT from commercially available instrumentation with a separate implementation of several new high-resolution signal and image processors imported from the sciences and engineering. However, many of the mathematically and technically different processors lead to spurious, i.e. unphysical resonances. Such unphysical peaks are often not easy to detect and regularize. These methods should be subjected to convergent validation, after which only the features of a spectrum/image that are faithfully reproduced by several processors could eventually be considered as candidates for reliable estimates. Other characteristics that fluctuate from one theory to another should be considered with caution. The cross verification of various methods should be done after signal encoding and, therefore, no changes are needed to the conventional scanning protocol. Hence, processing of the recorded signals could be done independently of scanners and this would lead to a greater flexibility in applying different spectral analysers. This could help, e.g. non-invasive diagnostics in medicine within the realm of MRS, MRI and MRSI, as clinicians would have more options than relying solely upon FFT images from commercial scanners. Needless to say, signal processing methods that have no recipe for regularization of spurious resonances are particularly undesirable in medical diagnostics. For example, in MRS the lactate and some lipid resonances usually have small intensities relative to the concentration of other metabolites and are, in fact, embedded in a large background. Spurious resonances could easily interlace with lipid peaks and cause considerable confusion in the identification pattern for diagnostic purposes.

The drawback of the mentioned convergent validation of different parametric estimators is that there is no guarantee that resonances reproduced by several methods are indeed the genuine ones. This is due to the intrinsic *instability* of all the existing parameter estimators. Therefore, alternative avenues should be explored, as done, e.g. in the present book, where a reliable procedure is designed to ensure stability of the PA by having to deal only with stable or Schur polynomials in the Padé numerator and denominator. This is the Padé–Schur approximant (PSA), with a rational polynomial ansatz which can be generated in any of the selected procedures including the Lanczos algorithm as elaborated in this book. A number of favourable capabilities (stability, robustness, efficiency, etc) of the PSA may prove to be well-suited for the task of adequately supplementing the FFT software in commercial scanners. The PSA might be conceived as an interface to new experiments in scattering and spectroscopy through signal/image processing with the possibility of reaching a substantially higher resolving power than the one currently available. Such an achievement could be exploited in the future to build new spectrometers that are based on, e.g. the PSA which also provides *en route* the conventional FFT spectra. To optimally focus on the region of interest, various segmentations of spectra and images could be done *a posteriori* along with resampling the signal at different sampling rates, but with preservation of the indigenous information in the selected region, etc.

Such a multi-faceted analysis is possible at the different equivalent acquisition times, but without having to repeat the experimental scanning itself. This high-resolution processing is aimed at extracting more information from measurements than is actually feasible by the FFT for a given fixed signal length. At first, this goal might seem paradoxical in view of the fact that the FFT is a linear transform which, therefore, must hold the same information as recorded in the measured time signal. However, this paradox is only apparent. Indeed the FFT does fully preserve the measured information, but a part of this contingent might be hidden in many Fourier spectra and images. This is due to a low resolution power of the FFT. Therefore the main task of any new high-resolution processor is to convert this obscure part of the FFT with its inaccessible information into the corresponding transparent information with potentially improved results.

One of the most reliable algorithms capable of providing the recommended, cross validated spectral information for the purpose of assisting, e.g. medical diagnostics is the FPT. This is the generic name for the PA applied to signal processing irrespective of which concrete computational algorithm is used to obtain the expansion coefficients of the Padé rational polynomial. In fact, the FPT is unique among all the existing signal processors, since it performs a self-contained cross validation. This stems from the fact that the FPT, as a system function designed to respond optimally to an external excitation, has two complementary variants. They are denoted by FPT^{+} and FPT^{-} to point at their expansion variables $z^{\pm 1}$ and to the two entirely different convergent regions located inside ($|z| < 1$) and outside ($|z| > 1$) the unit circle, respectively. Both FPT^{+} and FPT^{-} rely upon the same input spectrum given by the truncated and scaled Green function $\sum_{n=0}^{N-1} c_n z^{-n}$ where $z = \exp(i\omega\tau)$ with $\mathrm{Im}(\omega_k) > 0$, $\tau > 0$ and the set $\{c_n\}(0 \leq n \leq N-1)$ of the total length N represents the collection of all the encoded signal points. Here, the variant FPT^{-} is the standard diagonal PA given as the rational polynomial $A_K^{-}(z^{-1})/Q_K^{-}(z^{-1})$ in which both the input series development $\sum_n c_n z^{-n}$ and the PA have the same expansion variable z^{-1}. On the other hand, the FPT^{+} formally coincides with the so-called causal z-transform given by the diagonal PA via a rational polynomial in variable z, i.e. $A_K^{+}(z)/B_K^{+}(z)$ for the input sum $\sum_n c_n z^{-n}$, which is itself an expansion in powers of z^{-1}. The fundamental frequencies $\{\omega_k^{\mathrm{FPT}\pm}\}$, counted with their proper multiplicities (if any), are computed respectively via the $\mathrm{FPT}^{\pm}$ by rooting the characteristic equations $B_K^{\pm}(z^{\pm 1}) = 0$ in the complex z-plane[1]. Both variants $\mathrm{FPT}^{\pm}$ must give the identical spectral parameters that can jointly be denoted by $\{\omega_k^{\mathrm{FPT}}, d_k^{\mathrm{FPT}}\}$ and this occurrence stems from the uniqueness of the FPT for the same input sum $\sum_n c_n z^{-n}$. Then, by necessity, the only common location in the complex frequency plane, where the eigenvalues $\{\omega_k^{\mathrm{FPT}\pm}\}$ could possibly be very near each other $\omega_k^{\mathrm{FPT}+} \approx \omega_k^{\mathrm{FPT}-}$, is a segment located infinitesimally close to the unit circle, i.e. $\omega_k^{\mathrm{FPT}\pm} \approx \omega_k^{\mathrm{FPT}} \mp \varepsilon_k^{\mathrm{FPT}}$. Here, the quantity $\varepsilon_k^{\mathrm{FPT}} = \varepsilon_{k,\mathrm{R}}^{\mathrm{FPT}} + i\varepsilon_{k,\mathrm{I}}^{\mathrm{FPT}}$ is

[1] Hereafter, in order to avoid double superscripts, whenever the acronyms $\mathrm{FPT}^{\pm}$ are used in the superscript of a given quantity, say X, we shall write $X^{\mathrm{FPT}\pm}$ instead of $X^{\mathrm{FPT}^{\pm}}$.

an 'infinitesimally small complex number' which is defined by the corresponding pair of infinitesimally small real numbers $\varepsilon_{k,\mathrm{R}}^{\mathrm{FPT}} > 0$ and $\varepsilon_{k,\mathrm{I}}^{\mathrm{FPT}} > 0$. Thus, in principle, by selecting the accuracy thresholds $\varepsilon_{k,\mathrm{R}} > 0$ and $\varepsilon_{k,\mathrm{I}} > 0$ with $\varepsilon_k = \varepsilon_{k,\mathrm{R}} + i\varepsilon_{k,\mathrm{I}}$, the optimal frequencies $\omega_k \mp \varepsilon_k$ could be extracted with fidelity from those computed values $\{\omega_k^{\mathrm{FPT}\pm}\}$ that are infinitesimally different from each other $\omega_k^{\mathrm{FPT}\pm} \approx \omega_k^{\mathrm{FPT}} \mp \varepsilon_k^{\mathrm{FPT}}$ provided that $\varepsilon_{k,\mathrm{R}}^{\mathrm{FPT}} \leq \varepsilon_{k,\mathrm{R}}$ and $\varepsilon_{k,\mathrm{I}}^{\mathrm{FPT}} \leq \varepsilon_{k,\mathrm{I}}$. Moreover, in practice, we do not even need to guess the error estimates $\{\varepsilon_{k,\mathrm{R}}, \varepsilon_{k,\mathrm{I}}\}$. Instead, the sought genuine frequencies $\{\omega_k\} \approx \{\omega_k^{\mathrm{FPT}}\}$ will automatically be identified as those values $\{\omega_k^{\mathrm{FPT}+}\}$ that are equal to $\{\omega_k^{\mathrm{FPT}-}\}$ through a pre-assigned number of decimal places (say, two). Likewise, those frequencies $\{\omega_k^{\mathrm{FPT}\pm}\}$ that do not pass this stringent accuracy threshold are classified as spurious. Of course, this assumes that most of the computed frequencies and amplitudes $\{\omega_k^{\mathrm{FPT}\pm}, d_k^{\mathrm{FPT}\pm}\}$ in both variants have converged as a function of, e.g. the truncated signal length $N/M (M > 1)$. In this inherently controlled and self-contained manner, the FPT is able to simultaneously provide the genuine and identify the spurious resonances.

The power of this novel type of cross validation is in bypassing altogether comparisons among different methods that could yield approximately the same results, which nevertheless might turn out to be wrong. By contrast, the FPT^+ and FPT^- share a common conceptual design of a system response function via rational polynomials $A_K^+(z)/B_K^+(z)$ and $A_K^-(z^{-1})/B_K^-(z^{-1})$ within the same kind of a mathematical modelling, namely the FPT. Yet, the two variants FPT^+ and FPT^- are computationally so complementary to each other that they could rightly be considered as distinct strategies. This is because the FPT^- is an accelerator of a given slowly convergent series, whereas the FPT^+ induces/forces convergence into an initially divergent series by means of analytical continuation [28, 72, 75]. The FPT methodology accomplishes these two diametrically opposite tasks through its two complementary wings, the FPT^+ and FPT^- that converge inside $(|z| < 1)$ and outside $(|z| > 1)$ the unit circle, for the same input $\sum_n c_n z^{-n}$ which is itself divergent and convergent for $|z| < 1$ and $|z| > 1$, respectively. The optimal performances of the FPT^- and FPT^+ are achieved for $N = 2K$ and $N > 2K$, respectively, where K is the degree of the numerator and denominator Padé polynomials from the quotients $A_K^{\pm}(z^{\pm 1})/B_K^{\pm}(z^{\pm 1})$. The cases $K = N/2$ and $N > 2K$ correspond respectively to an algebraically determined and over-determined system of linear equations for obtaining the expansion coefficients of the polynomials $A_K^{\pm}(z^{\pm 1})$ and $B_K^{\pm}(z^{\pm 1})$. These different systems of linear equations encountered in the FPT^+ and FPT^- employ the same input information, i.e. truncated and scaled Green function $\sum_{n=0}^{N-1} c_n z^{-n}$.

Notice that for the purpose of illustration, the above highlights focused on explaining the Padé self-consistent cross validation mainly for complex frequencies $\{\omega_k^{\mathrm{FPT}\pm}\}$. However, the same type of stringent accuracy thresholds are used also for cross validation of the associated complex amplitudes $\{d_k^{\mathrm{FPT}\pm}\}$ that are computed from their analytical expressions in the $\mathrm{FPT}^{\pm}$. As the final

results, we obtain the spectral parameters $\{\omega_k^{\mathrm{FPT}}, d_k^{\mathrm{FPT}}\}$ that are the intersection of the two computed sets $\{\omega_k^{\mathrm{FPT}+}, d_k^{\mathrm{FPT}+}\}$ and $\{\omega_k^{\mathrm{FPT}-}, d_k^{\mathrm{FPT}-}\}$. Once the sought spectral pairs $\{\omega_k^{\mathrm{FPT}}, d_k^{\mathrm{FPT}}\}$ are faithfully extracted from the encoded data $\{c_n\}$, the Padé complex mode shape spectra (called hereafter the Padé parametric shape spectra) given by the Heaviside partial fractions [132]–[141] could be at once constructed. These latter shape spectra determined from the estimated pairs of parameters $\{\omega_k^{\mathrm{FPT}\pm}, d_k^{\mathrm{FPT}\pm}\}$ must, up to random noise differences, coincide with the corresponding complex mode shape spectra (called hereafter the Padé non-parametric shape spectra) computed using only the quotients $A_K^{\pm}(z^{\pm 1})/B_K^{\pm}(z^{\pm 1})$ which do not rely at all upon the complex frequencies and amplitudes $\{\omega_k^{\mathrm{FPT}\pm}, d_k^{\mathrm{FPT}\pm}\}$. Any difference between the Padé parametric and non-parametric shape spectra detected above the signal-to-noise ratio (determined as, e.g. the rms—the root mean square of the complex mode FFT spectrum) would indicate that the estimated parameters $\{\omega_k^{\mathrm{FPT}\pm}, d_k^{\mathrm{FPT}\pm}\}$ need to be refined via an appropriate optimization. We accomplish this optimization via an extremely powerful, iterative, self-correcting and stable algorithm based on the Maclaurin expansion for the spectral parameters, by minimizing the squared difference between the Padé parametric and non-parametric shape spectra within the same variant, e.g. the FPT^+ [142]. Of course, we proceed likewise with this kind of optimization while using the FPT^-. The initial values of such two separate and independent iterative optimizations in $\mathrm{FPT}^{\pm}$ are the spectral pairs $\{\omega_k^{\mathrm{FPT}\pm}, d_k^{\mathrm{FPT}\pm}\}$ that stem from the application of the explained parametric versions of the $\mathrm{FPT}^{\pm}$. The iterations are stopped when the difference between the Padé parametric and non-parametric shape spectra is indistinguishable from the random noise, or equivalently, the computed rms. The output spectral parameters, denoted again for simplicity by the same labels as the initial values $\{\omega_k^{\mathrm{FPT}\pm}, d_k^{\mathrm{FPT}\pm}\}$ are optimal in the mentioned least-square sense. This constitutes an internal validation of the two sets of estimates $\{\omega_k^{\mathrm{FPT}+}, d_k^{\mathrm{FPT}+}\}$ and $\{\omega_k^{\mathrm{FPT}-}, d_k^{\mathrm{FPT}-}\}$ in an entirely independent manner without any direct reference of one set of spectral parameters from the FPT^+ to the other from the FPT^-. It is only after accomplishing such intrinsic validations of parameters $\{\omega_k^{\mathrm{FPT}+}, d_k^{\mathrm{FPT}+}\}$ and $\{\omega_k^{\mathrm{FPT}-}, d_k^{\mathrm{FPT}-}\}$ that we proceed towards the critical cross validation by identifying the intersection between these latter two sets with the emergence of the final estimates $\{\omega_k^{\mathrm{FPT}}, d_k^{\mathrm{FPT}}\}$ for an accurate and maximally reliably quantification of the unknown exact physical resonances $\{\omega_k, d_k\}$.

Overall, there is a strong need for concepts and methods from quantum mechanics and quantum scattering theory [143]–[157] in many basic sciences other than physics, as well as in applied sciences, including life sciences and beyond. This is not just because quantum mechanics is a complete physics concept, which is among the best established strategies in all sciences, thoroughly validated in innumerable comparisons with high-precision experimental measurements that provide the most stringent tests, it is also because the versatile mathematical tools of this theory are well-suited for direct

implementations in other fields. Even when similar tools are used elsewhere, as indeed they are not necessarily invented in quantum mechanics, but rather in pure and/or applied mathematics, a rich research experience gained in quantum-mechanical complicated applications and sophisticated computations could be highly beneficial to research beyond physics. A partial account on this point is given in the present book, which illuminates the quantum-mechanical tight binding model or, equivalently the nearest neighbour approximation [96, 97], using spectral analysis based upon the Lanczos recursive algorithm [93]–[104]. Working along these latter lines as well as intertwining the Lanczos and Padé methods in the setting of the tight bindings, we have recently [17, 18] derived the practical and general exact analytical formulae for the following key quantities: (i) the expansion coefficients $\{a_n\}$ of the continued fractions (CF), (ii) the Lanczos coupling parameters $\{\alpha_n, \beta_n\}$, (iii) the Lanczos polynomials $\{Q_n(u), P_n(u)\}$, (iv) the fast Padé transform $(c_0/\beta_1)P_n(u)/Q_n(u)$, (v) the Lanczos state vectors ψ_n, (vi) the total wavefunction $\Upsilon(u)$ of the studied general system at any complex variable u and (vii) the 'Hamiltonian' operator $\hat{\Omega}$. All the closed formulae (i)–(vii) rely only upon the given input data of experimentally measured time signals $\{c_n\}$ or theoretically generated auto-correlation functions $\{C_n\}$ of arbitrary length N. Moreover, a multi-dimensional extension of the FPT has also been formulated and implemented to single, double and triple quadratures [157, 158] with an unprecedented accuracy outperforming spectacularly the FFT by ten decimal places. In [24], the bivariate FPT has been devised and successfully applied to two-dimensional magnetic resonance spectroscopy (2D-MRS) for cross correlation plots using biomedical time signals $\{c_{n_1,n_2}\}$ encoded from patients. The first application of the FPT to phantom and volunteer data from MRI reported in [34] also shows great promise especially for reducing the truncation artefacts known as the Gibbs oscillations.

The spectral analysis or frequency analysis problem has a long history which can be traced back to the harmonic inversion problem of Prony [57]. This problem is a decomposition of a given function into its components comprised of complex harmonics whose parameters are unknown. It is stated as follows: given a set of input data, via, e.g. time signal points $\{c_n\}$ or auto-correlation functions $\{C_n\}$, one is required to find the number of the constituent resonances, their complex positions and the corresponding residues. This is recognized as one of the main topics in the quantum-mechanical resonant scattering theory and spectroscopy that are among the chief physics methods for studying the structure of matter on different levels and scales [143]–[157]. This observation alone could justify the effort in attempting to tie together signal processing with quantum-mechanical resonance scattering and spectroscopy [108]–[116].

An apparently different subject, the power moment problem from celestial mechanics is set up to search for the mass distribution/concentration on different locations, i.e. spatial positions in a given constellation or a system or a body for the known total mass [159, 160]. However, this is mathematically equivalent to the harmonic inversion in processing time signals and, in particular, to mass

spectroscopy in ICR in analytical chemistry or to MRS in medical diagnostics of human tissue. The method of the power moment is not frequently studied in quantum mechanics [161]. By contrast, this method is abundantly used in mathematical probability theory and in statistics [162]–[175], whose results could give a qualitatively new insight into ordinary or generalized quantum-mechanical eigenvalue problems as well as into the harmonic inversion in signal processing and the like. For example, the characteristic polynomials, i.e. eigenpolynomials that originate from the given secular equation, as the building blocks of a number of spectral analysers, are known to be exponentially ill-conditioned when constructed using the power moments. The situation can be dramatically improved when the power moments are weighted with one [162]–[170] or two [171]–[175] orthogonal polynomials that lead to the modified and mixed moments, respectively. This is analysed in chapters 53–56. In the case of the mixed moments, one of the two polynomials is given in advance and, as such, could be advantageously selected from the family of the known, classical orthogonal polynomials (Legendre, Laguerre, Lagrange, Hermite, Chebyshev, Gegenbauer, Neumann, etc) [176]–[180]. The mixed moments enable an extremely accurate and stable construction of the needed characteristic polynomials of very high orders. This numerically robust method, especially in the formulation of Wheeler [174], is capable of producing the complete set of the coupling parameters $\{\alpha_n, \beta_n\}$ that are encountered in the three-term contiguous recursions for polynomials[2]. This capability yields high precision which is achieved systematically within machine accuracy, as has been demonstrated by Gautschi [175] for moments defined at finite integration limits.

It has been stated in *Numerical Recipes* [176] that the power moment is notoriously ill-conditioned and, as such, should not be used for computations. Indeed, the computations of Gautschi [175] based on the power moments shows that the coupling constants $\{\alpha_n, \beta_n\}$ can lose all their significant digits for the values of n that are of a relatively low order, e.g. 10–15. However, Gordon [170] also used the power moments in a computation of the constants $\{\alpha_n, \beta_n\}$ via the product-difference (PD) recursive algorithm and arrived at machine accurate results. Therefore, it is not the use of power moments which should be abandoned, but rather the way in which they are incorporated into the subsequent generation of the parameters $\{\alpha_n, \beta_n\}$. The unprecedentedly powerful PD algorithm of Gordon [170] was published in 1968, but this seminal work passed unnoticed in the mentioned papers of Wheeler [174] from 1974 and Gautschi [175] from 1978. The PD algorithm is similar to the more familiar quotient-difference (QD) algorithm from 1957 attributed to Rutishauser [169], although it was known to Frobenius in 1879 [66], as pointed out previously by Wynn [73]. The PD algorithm performs the division only once at the end of the recursion. However,

[2] The same constants $\{\alpha_n, \beta_n\}$ are also encountered in the Lanczos algorithm [93]–[104] for the state vectors $\{\psi_n\}$.

the QD algorithm uses divisions in each iteration and in a finite-precision arithmethic this could lead to more round-off errors than in the PD algorithm.

The Lanczos polynomials that emerge from the algorithms of [170] and [174] are expected to preserve orthonormality as well as the *linear independence* throughout the computation for high orders. Once the coupling constants are generated, and the polynomials constructed, a *complete* basis set of the expansion functions with conserved orthonormalization becomes automatically available. No numerical quadrature is required for the generation of the key ingredient of the method, namely the coupling parameters $\{\alpha_n, \beta_n\}$. The methods of Gragg [171] and Wheeler [174] for the modified and mixed moments, respectively, have their roots in the original Chebyshev [162] algorithm. The mentioned demonstrations by Gordon [170] and Gautschi [175] of the performances of the PD and the Wheeler algorithms, respectively, should be contrasted to one of the most frequently employed methods in quantum chemistry and in solid state physics, i.e. the Lanczos recursive algorithm [93]–[104], which generates the state vectors that are observed to suffer from severe numerical instabilities in striving to maintain orthogonalization and linear independence. An accurate and stable extraction of all the required coupling parameters $\{\alpha_n, \beta_n\}$ and the associated expansion basis set functions from the mixed moment problem are of direct use in versatile applications, e.g. solving multi-channel Schrödinger eigenvalue problems in quantum mechanics, scattering problems, signal processing, etc. For example, the constants $\{\alpha_n, \beta_n\}$ can instantaneously provide the resolvent or the Green functions, Lanczos continued fractions (LCF) and the Padé–Lanczos approximant (PLA) [19, 85, 97, 120]. These three mathematically equivalent methods can yield the complete spectral information (eigenenergies, eigenfunctions, residues) by rooting the characteristic polynomials. This is the parametric aspect of the mentioned estimators. Alternatively, they could provide shape spectra without rooting the characteristic polynomials and this role is reminiscent of transforms in the spirit of the Fourier transform. Moreover, we shall demonstrate in the present book that the coupling parameters $\{\alpha_n, \beta_n\}$ are also sufficient for a complete construction of auto-correlation functions $\{C_n\}$ without the knowledge of either u_k or d_k [19]. This is in sharp contrast with the current practice, which necessitates the entire collection of the eigenvalues and residues $\{u_k, d_k\}_{k=1}^{K}$ for computations of auto-correlation functions [181].

Root searching is a nonlinear procedure which represents a bottleneck of all spectral methods based upon polynomials. Therefore, a direct rooting of high-order polynomials should be avoided whenever possible. Fortunately, this is feasible, since finding all the roots of any polynomial is completely equivalent to diagonalization of the corresponding Hessenberg matrix [176]–[179]. This latter matrix is extraordinarily sparse, since it has mainly zeros throughout its structure, except in one of the diagonals with all the elements equal to unity and in the first row containing the expansion coefficients of the characteristic polynomial. Diagonalization of such a matrix is computationally manageable even for a very large dimension and, therefore, root search of polynomials can

be completely bypassed, i.e. replaced by an equivalent diagonalization of the associated Hessenberg matrix, as has recently be done in signal processing [115]. All told, a versatile and universal role of non-classical orthogonal polynomials becomes exceedingly attractive when established with the help of the stable, accurate and recursive algorithms of Gordon [170] and Wheeler [174] for moment problems. The unprecedented success of these two algorithms within the moment problem are indeed invaluable and their full exploration in physics and chemistry as well as in signal processing and beyond is yet to come. Alternative and/or complementary to the numerical algorithms from [170] and [174], the analytical/closed and simple (i.e. non-determinantal) formulae have been reported in [17] for the Lanczos coupling constants $\{\alpha_n, \beta_n\}$ in the general case of an arbitrary value of the non-negative integer n. This implies that, e.g. the resulting shape spectra are also available from an analytical formula in the form of a finite-order continued fraction which is defined solely in terms of the constants $\{\alpha_n, \beta_n\}$.

Recall here that continued fractions are among the most stable and most robust computational tools from numerical analysis as well as from theory and practice of approximations of functions [182]–[192]. This statement is readily substantiated by merely a cursory inspection of any computer main frame and/or the leading mathematical software libraries for computations of nearly every elementary function (exponentials [187], trigonometric functions [188], etc) and special functions (Bessel functions, gamma-function, probability/error function [189, 190], Gauss hypergemetric functions [183], Coulomb regular/irregular functions [192], etc) [176]–[179]. For example, a computation of the exponential $\exp(-x)$ for e.g. $x = 20$ should, at first sight, appear as trivial through the corresponding Maclaurin expansion which converges for every x. Nevertheless, the results obtained in this manner are entirely unsatisfactory, as opposed to the corresponding continued fractions which work perfectly well for any x including $x = 20$ from this particular example [193].

One of the goals of the present book is to carry out spectral analysis from the viewpoint of the power, modified and mixed moment problems. This is particularly motivated by the accuracy, stability and efficiency of the algorithms of Gordon [170] and Wheeler [174] for the power and mixed moment problem. On top of this, we add the present analytical formula for the coupling parameters $\{\alpha_n, \beta_n\}$. The advantages of these three methods are expected to lead to significant improvements in harmonic inversion, as well as in obtaining complete spectral information for any given Hamiltonian or other dynamical operators in chemistry, physics and beyond. This is highly relevant to applications across MR physics and especially in MRS. For example, the so-called quantification problem in MRS, i.e. determining concentrations, relaxation times and chemical shifts of metabolites in a given tissue of the human body is entirely equivalent to the moment problem, and yet this line of thought remains virtually unused in applications within MR physics. This could be achieved through a number of accurate, stable and simple algorithms analysed in the present book.

Among the most commonly used nonlinear *fittings* for processing *in vivo* FIDs recorded within MRS is the so-called LCModel [11]. This acronym stands for the linear combination of model *in vitro* spectra from individual metabolite solutions. In the preprocessing stage, the LCModel usually doubles the number of data points in the time and frequency domains by using the zero-filling in both *in vitro* and *in vivo* data. If a spectrum in the frequency domain is approximated by a sum of Gaussians or Lorentzians, then the corresponding form of the FID in the time domain will be given by a linear combination of damped exponentials. In such a case, any fittings in the time and frequency domains are necessarily nonlinear. This should be contrasted to linear mappings, such as FFT, in which the fundamental Fourier frequencies in the exponentials are fixed prior to processing. Nonlinear fits are iterative and some initial values are needed for all the freely fitted parameters of a chosen mathematical model. Since these values are unknown prior to fitting, they must be either guessed or produced by an auxiliary mathematical model or acquired via a separate measurement of FIDs in MRS on, e.g. some related phantoms. The latter alternative is adopted in the LCModel where the initial values for a nonlinear fit are taken from data banks or libraries of those *in vitro* measured spectra that correspond to the examined *in vivo* spectrum. Here, it is assumed that the measuring instrument, magnetic field and the encoding protocol/sequence are the same for acquiring both the *in vitro* and *in vivo* spectra. The LCModel does not assume *a priori* that the measured *in vitro* and *in vivo* spectra are of the Lorentzian or Gaussian type or of any other pre-assigned form. However, an overall exponential factor of the model for the complex spectrum is dependent upon two unknown phase corrections that are treated as the fitting parameters of the LCModel. This implies that the LCModel is nonlinear even if everything else were dependent upon the unknown parameters in a linear way. Prior to using the LCModel in each laboratory, one must measure a set of the *in vitro* spectra on a selected phantom, which should match as closely as possible the human or animal organ whose *in vivo* spectrum will be subsequently subjected to signal processing. Then a mathematical model is set up as a linear combination of these measured *in vitro* spectra from metabolite solutions. The unknown constants in such a sum are the metabolite concentrations, phases, line broadening shifts, line shapes and baseline polynomial coefficients. The latter coefficients are used to construct a cubic B-spline piece-wise polynomial for description of the background contribution to the given spectrum. Resonances are superimposed on top of this rolling baseline of the spectrum. The above sum of the *in vitro* model spectra is fitted to the given *in vivo* spectrum by using the nonlinear Levenberg–Marquardt [194] iterative algorithm with the Newton–Gauss type of a constrained least-square minimization. The final results of this fitting are the *in vivo* spectral parameters that were the mentioned unknown constants of the LCModel.

The common limitation of all nonlinear fits is *non-uniqueness*. This is to say that, within the same prescribed uncertainty, a measured spectral structure could be fitted by, e.g. two, three or four shape lines (Gaussians and/or Lorentzian, etc), without a sound criterion determining which of the fits should be retained in the

end of the analysis. Such a lack of a criterion of established validity could readily yield spurious metabolites (over-fitting) or cause some genuine metabolites to pass undetected (under-fitting). Either outcome could lead to difficulties that would inevitably limit diagnostics. The capabilities of the LCModel are in the absence of assumptions about any pre-assigned line shape of the studied spectrum and also in the usage of the Levenberg–Marquardt nonlinear optimization for determination of the peak parameters. One of the limitations of the LCModel is that it necessitates the prior information on model *in vitro* spectra from data bases acquired by a subsidiary measurement on the related phantom. If the existing data banks do not contain the needed model spectra, supplementary encoding is necessary for the purpose of initiating the LCModel. Rather than carrying out these additional measurements on phantoms, Monte Carlo simulations have occasionally been employed [12, 13] for creating data banks that would provide the Levenberg–Marquardt algorithm with alternative sources of the initial spectra. In the LCModel the number of metabolites N_M is fixed in advance and it is usually set to be equal to 15 or so. It could be better to consider this number N_M as one of the unknown parameters of the problem. A suitable pre-assigned number N_{M_0} could play the role of the initial value, in which case the final number N_M would come out from the list of the optimized parameters from the Levenberg–Marquardt algorithm. Even with this potential refinement which has not been implemented in the literature thus far, the ensuing number N_M of estimated metabolites could at best be only a rough approximation, which is controlled solely by the appearance of the FFT shape spectrum, rather than the genuine information hidden in the measured time signal. By contrast, this true information about the exact number of resonances or metabolites is extractable directly from the raw FID by one of the variants of the Shanks transform [72] which belongs to the Padé methodology [19]. Practice has shown that removing, e.g. only one metabolite from an *in vitro* spectra leads to poor fits in a part of the *in vivo* spectrum where the missing model resonance normally occurs [11]. An obscure interpretation for this latter situation has been put forward stating that 'there should be no reason for such a removal, since generally a metabolite which has a good possibility of being detected should be included in the basis set' [11]. This interpretation itself is at variance with a proclaimed objectivity of the LCModel [11]. In reality, what one hopes to obtain from an *in vivo* spectrum by using the LCModel is directly proportional to what one puts by hand into the basis set through *in vitro* model spectra as has recently been emphasized [26]. Hence subjectivity of the LCModel. Even in normal tissue there is a noticeable degree of variation of metabolite concentrations that could roughly be estimated from the product of the peak hight and peak width. In tumorous tissue some of the metabolite resonances might be unidentifiable from a spectrum, since they could be buried in noise or found at some other shifted positions or detected with heights and widths having quite different values from those in normal tissues. This is especially true for the two lactate resonances that are usually difficult to see due to a large background in spectra of the brain gray matter

measured at a long echo time (TE), e.g. TE = 136 ms or TE = 272 ms as analysed in [14]. In such a case, no spectral fitting techniques would be adequate, including the LCModel. Lactate and lipid resonances are clustered together, but the relaxation times of the latter are shorter than those of the former. Hence, using larger values of TE, as alluded to above, appears to be better, since it would allow the lipid resonances to decay and this should facilitate the identification of the lactate peaks. On the other hand, a larger TE would enter the tail of the FID envelope where the the signal is weak and, as such, comparable with the intensities of random noise. No fitting of FFT spectra of this type should be expected to give any reliable results. Therefore, an alternative to fitting of low-resolution FFT spectra is sought. Such an alternative must be capable of identifying even the weakest resonances and, e.g. the Padé-type methods are well suited for this purpose [27]–[29]. As already mentioned, in a normal spectrum of the healthy brain gray matter, the lactate and lipid resonances are practically unrecognizable from the background. However, lactate elevations are discovered in a number of brain disorders, e.g. ischemia, progressive leukodystrophies, severe inflammation, malignancies, etc. Lipid enhancements are also often related to tumours, multiple sclerosis plaques, etc. Precise theoretical estimators of all the peak parameters of other resonances are also needed in helping diagnostics in the case of data recorded at short values of TE. For example, at TE $\sim$ 20 ms, the detection of a mere decrease of the height of the NAA (Nitrogen–Acetyl–Aspartate) metabolite, which is the strongest resonance after the giant water peak, could be diagnostically important. Such an indicator from cerebral proton MRS spectra could be advantageously available before any pathological alteration has been observed on the corresponding anatomical scans from MRI.

The field of MRS needs a more vigorous access to methods that are alternative to fitting devices and these can be found within a number of parametric estimators of spectra [108]–[116]. One such estimator is the PSA [18] which can be used both as a parametric and/or spectral processor. The parametric version of the PSA could be employed in, e.g. MRS which aims at quantification of spectra, yielding the precise quantitative characteristics of the principal resonances via determination of their positions, widths, heights, phase, relaxation times, etc. The non-parametric version of the PSA is pertinent to, e.g. MRI where one is not interested in evaluating the numerical values of the reconstructed features, but rather attention is focused onto the anatomy of the imaged organ of the human body. There is also a newly emerging hybrid discipline MRSI. Here, one is interested in topological structures of the imaged subject in a chosen segment as well as in quantitative evaluations of the purely spectroscopic type of the selected slices. Both of these important diagnostic tasks in current medical practice could be successfully accomplished by the application of, e.g. the mentioned two versions of the PSA. The DPA [9, 115] is also a parametric and a non-parametric method, but it cannot cover the whole Nyquist range in one run, since it relies upon windowing, as do the FD, DSD or DLP processors. The common price paid by FD, DSD, DPA and DLP methods for their achieved

dimensionality reduction of large data matrices associated with long signals is limitation to only local spectral analysis in one run. The PSA uses no windowing or decimation and, therefore, both local and global spectral analyses are possible in a single application of the method without patching together the separate frequency intervals. These four methods (FD, DSD, DLP and DPA) refer to windowed, i.e. local spectra in a selected frequency interval $[\omega_{\min}, \omega_{\max}]$ which is a part of the the full Nyquist range $[-\pi/\tau, +\pi/\tau]$. Every type of windowing in either the time or frequency domain ignores the possible effects of frequencies outside the chosen local range $[\omega_{\min}, \omega_{\max}]$ onto the informational content within this interval of interest. Such effects exist especially for *in vivo* signals encoded by e.g. proton MRS from human brain where detection of dilute metabolites is required in the presence of a giant water resonance which is about 10^4 times larger than other peaks in the corresponding frequency spectrum. To cover the entire Nyquist range, the windowing methods perform the spectral analysis window-by-window, such that all the local spectra are afterwards glued together in the consecutive order to yield the complete spectrum. This window patching often leads to some unwanted edge effects requiring special care to avoid artefacts, e.g. aliasing phenomena, the Gibbs oscillations and the like [64]. Such occurrences are enhanced for overly narrow frequency intervals imposed by exceedingly long time signals and it is, therefore, desirable to have a generic method, such as PSA, which does not need to resort to windowing.

The experimental resolution power of many instruments such as spectrometers is limited in part by a particular theory, which is the FFT. This method should be complemented by other more powerful high-resolution parametric estimators. The latter processors are able to extract, directly from the measurements of $\{c_n\}$, without any post-processing fits, the main spectral features, i.e. information about resonances. For example, in the parametric version of the PSA, the final outcome is a set of tabular numerical values of the peak parameters of each resonance found in the analysis using only the raw signal points $\{c_n\}$. This provides an alternative to fitting procedures that are customarily in use within MRS. When dealing with conventional spectrometers, one is usually unaccustomed to such tables of spectral results, since the first thing which is viewed directly on the screen is the magnitude or power spectrum. In such a case, if the peak parameters are required, one would fit each of the FFT peaks to a pre-assigned line shape comprised of, e.g. one or more Lorentzian and/or Gaussians[3]. As emphasized earlier, such a standard post-processing analysis requires prior information, which is the knowledge of the initial values of the parameters for each individual resonance before undertaking the task of the peak parameter search. By contrast, the PSA does not necessitate any prior knowledge, since the fundamental frequencies *as well as their total number*, and the corresponding amplitudes, are extracted directly from the raw signal. Of course, the peak

[3] One of the alternatives to this peak-by-peak fit would be adjustments of a linear combination of a premeasured sequence of spectra to a studied spectrum as done, e.g. in the LCModel [11].

parameters obtained in such a computation can be used to construct spectra in various modes, e.g. magnitude, power, absorption, etc. This change of strategy via 'extracting first all the relevant peak parameters from the signal and then constructing any desired spectrum', constitutes a fundamental advantage of the class of parametric methods [108]–[116], relative to the standard Fourier-based spectroscopy and the accompanying nonlinear fittings [11, 194]. Using various initial guesses while post-processing FFT spectra through a nonlinear fitting, could produce significantly different final outcomes that might deviate from the expected exact results. Furthermore, often very complicated and congested spectra with tightly packed peaks and overlapping resonances are encountered in MRS, hampering reliable fittings and this reduces fidelity in the whole processing via adjustable parameters. Such fittings become particularly difficult for degenerate or quasi-degenerate spectra, where several resonances can emerge with nearly equal frequencies, as is actually the case with a number of molecules[4]. Moreover, the literature witnessed no attempts thus far by the LCModel aimed at quantification of 2D-MRS cross-correlation plots which, by contrast, have recently been successfully quantified within the Padé method [24]. These listed drawbacks of the LCModel are clearly absent from the PSA which invokes no adjustable quantities whatsoever and, therefore, requires no initial values for the sought peak parameters. Of course, this is also the case with other parametric, non-iterative estimators, e.g. the FD [108]–[112], DSD, DPA or DLP [115], but the present emphasis is on the Padé methodology due to its unique reliability in handling the spurious roots entirely by analytical means, i.e. not through some numerical procedures. This is operationalized by one of the Padé algorithms, the already mentioned PSA, which can describe both Lorentzian (non-degenerate) and non-Lorentzian (degenerate) spectra. Experimentally measured spectral profiles could behave like the Gauss or the Voigt distribution functions [195]–[202]. It should be pointed out that it is possible to extend the PSA and encompass the Gauss and/or Voigt shape functions [142].

In the present illustrations, the Padé approximant is used for an accurate quantification of proton 1D-MRS spectra of the occipital gray matter of the human brain. The corresponding FID have been measured at the magnetic field strengths of 7T [13]. We obtain maximally accurate peak parameter values by relying only upon the measured raw FID with no other prior information and without zero-filling, preprocessing or post-processing the data [16]–[29]. The baseline of the FFT-MRS spectrum is considerable [13]. Nevertheless, we do not resort to any separate modelling for the background to account for this baseline, but nevertheless the complete spectral information is unfolded from the experimentally measured FID. *Before* drawing a graph to construct the shape of the investigated spectrum, all the metabolites are unequivocally identified with their corresponding peak parameters for every individual resonance. The total number of resonances is also one of the outcomes of the analysis within the

[4] An example of a molecule with a quasi-degenerate spectrum is CO_2.

Padé approximant. This number is determined exactly by a precise determinantal (Hankel) condition from the system of linear equations encountered in the theoretical development.

Chapter 2

Auto-correlation functions

In the present book, we focus on a unified theoretical treatment of collisions and spectroscopy using the concept of the auto-correlation function $C(t) = (\Phi(0)|\Phi(t))$ with a natural link to signal processing. From the onset, this strategy is rooted in basic quantum mechanics [143, 144] and quantum-scattering theory [145]–[157], since the state vector $|\Phi(t))$ is the solution of the time-dependent Schrödinger equation[1] $\hat{\Omega}|\Phi(t)) = i(\partial/\partial t)|\Phi(t))$. In quantum mechanics of genuine bound states and a pure continuum, the dynamics of a considered physical system is described by a Hamiltonian $\hat{\mathrm{H}}$, which as a Hermitean operator $\hat{\mathrm{H}}^{\dagger} = \hat{\mathrm{H}}$ coincides with $\hat{\Omega}$ in the above Schrödinger equation for $|\Phi(t))$. The eigenvalues of a Hermitean operator are real. However, to include resonances via complex energy eigenvalues, as the most important part of scattering, one can generalize the notion of a 'Hamiltonian' and extend it to encompass a non-Hermitean dynamic operator $\hat{\Omega}$ in the same Schrödinger formalism. Hereafter, non-Hermiticity of $\hat{\Omega}$ might imply that the scalar product in the vector space $\mathcal{H}$ is defined as the symmetric inner product $(\zeta|\xi) = (\xi|\zeta)$. No conjugation via the star superscript is placed onto either of the two state vectors or 'orbitals' $(\zeta|$ or $|\xi)$ that both belong to $\mathcal{H}$. To symbolically indicate this special feature of the symmetry of the scalar product, soft round brackets $|\cdots)$ and $(\cdots|$ are used in place of the usual Dirac 'bra-ket' notation $\langle\cdots|$ and $|\cdots\rangle$ with $\langle\zeta|\xi\rangle = \langle\xi^*|\zeta^*\rangle$ and $\langle\zeta^*|\xi\rangle = (\zeta|\xi)$. An abstract formulation of quantum mechanics and its deterministic postulate imply that, if the wave packet $|\Phi(0))$ of *any generic system* under study is well prepared/controlled at

[1] As Pauli [203] remarked, since relativity and quantum mechanics should be among the hypotheses of every adequate physics theory, it is plausible to redefine their respective basic quantities c and $\hbar = h/(2\pi)$ as unity, where c is the speed of light in vacuum and h is the Planck constant. In the ensuing so-called natural units ($c = 1 = \hbar$) all other physical quantities will have the dimensions of a power of length. If such reduced dimensions are multiplied by the requisite powers of c and $\hbar$, the true dimensions of the given quantity would follow. For example, in the natural units, the energy E has the dimension cm^{-1}. However, in reality, the energy divided by $c\hbar$ is a quantity with the dimension cm^{-1}. Throughout this book, unless otherwise stated, the natural units (also called the atomic units) $c = 1 = \hbar$ will be employed. This is convenient especially for the present analysis, since we can interchangeably use the frequency (ω) and energy (E) as two synonyms, $E = \hbar\omega = \omega$.

the initial time $t = 0$, and if its further development is propagated by the given dynamics/interactions, then the state $|\Phi(t))$ will be known exactly at any later instant t [157]. The state $|\Phi(t))$ is an element of the abstract vector space $\mathcal{H}$. Since $|\Phi(t))$ is used to derive $C(t)$, it follows that this latter quantity also represents an abstract concept. This in itself means that the auto-correlation functions $C(t)$ are independent of the origin from which they are generated and, therefore, could be computed theoretically or measured experimentally as, e.g. time signals $c(t)$. Moreover, the mathematical equivalences $C(t) = c(t)$ and $C_n = c_n$ exist for both the continuous (t) and discrete $(t = t_n \equiv n\Delta t)$ variables[2], if a given time signal is a sum of damped (attenuated) complex exponentials that yield a spectrum as a linear combination of pure Lorentzians [108]–[115]. In either case, the auto-correlation functions $C(t)$ or the time signals $c(t)$ physically represent the instantaneous *survival probability amplitude* of the corresponding time-dependent state or wave packet $|\Phi(t))$ of the examined system. This is important for at least two reasons: (1) experimental raw signals $c(t)$ can be used directly, without necessarily relying upon the theory, to deduce *by computations* the basic observables for scattering and spectroscopy, such as cross sections, rate coefficients or the like, and (2) measured time signals $c(t)$ that are also identifiable as counts per channel, can be directly and dynamically intertwined with the theory on a deeper fundamental level to yield more valuable spectral information.

It should be emphasized that the concept of auto-correlation functions first emerged as a computational tool, but soon afterwards surpassed its initial purpose. This is because the auto-correlation functions represent a veritable alternative formulation of quantum mechanics and quantum-mechanical resonant scattering theory. Many of the major observables, e.g. complete energy spectra, local density of states, quantal rate constants and other related quantities are expressible through auto-correlation functions or their suitable transforms. Other important observables could be given completely in terms of some appropriate, relatively small informational parts that might be singled out and analysed separately from the unwanted/redundant remainder of the full data set of auto-correlation functions. In order to theoretically generate auto-correlation functions $C(t)$, that otherwise play a critical role in analysis of spectra of physical systems, all the parametric methods require that the peak parameters $\{\omega_k, d_k\}$ are computed first as pairs of complex numbers. These are the fundamental frequencies $\{\omega_k\}$ and the associated residues $\{d_k\}$ that constitute the natural harmonics whose linear combination of K terms represents the building block of every individual time signal point c_n from the set $\{c_n\}$. Here, the elements of the triple $\{\mathrm{Re}(\omega_k), \mathrm{Im}(\omega_k), |d_k|\}$ are the position, width and height of the kth peak/resonance. One can use either the dynamic matrix $\mathbf{\Omega}$ or the related evolution/relaxation matrix $\mathbf{U} = \exp(-i\mathbf{\Omega}\tau)$ to generate the corresponding Jacobi matrix $\mathbf{J}$ whose diagonalization [176]–[179] can give the required spectral

[2] Here, $C_n \equiv C(n\tau)$ and $c_n \equiv c(n\tau)$, where $\tau = \Delta t$ is the sampling time, i.e. the segment between any two equidistantly sampled points on the real time axis t.

set $\{\omega_k, d_k\}$. However, this is not the only way as, in fact, the non-parametric version of the FPT is *also* able to generate any auto-correlation function C_n [18]. Such a version of the FPT does not compute the pair $\{\omega_k, d_k\}$ at all. Instead, it merely generates the general expansion coefficients $\{a_n\}$ in the continued fractions (CF) of any finite order. Nevertheless, it has been shown [18] that this is fully sufficient for the exact determination of the auto-correlation functions $\{C_n\}$ *without ever rooting* the characteristic equation $Q_K(\omega) = 0$, which otherwise gives the eigenvalues $\{\omega_k\}$ of $\hat{\Omega}$. Our procedure represents an important advantage relative to the current practice in many parametric estimators, e.g. the FD [108]–[112], the DSD, DLP or DPA [113]–[115], the Lanczos algorithm [181] where the eigenvalues and residues $\{\omega_k, d_k\}$ are determined first before constructing the auto-correlation functions $\{C_n\}$.

Chapter 3

The time-independent Schrödinger equation

Irrespective of whether one is concerned with spectroscopy or collisions, the whole physics of any given system is ingrained in one single quantity, which is the full Green function $(\zeta|\hat{\mathrm{G}}(\omega)|\xi)$ in a given representation $\{(\zeta|, |\xi)\}$ with $\hat{\mathrm{G}}(\omega)$ being a resolvent known as the Green operator [145]–[157]

$$\hat{\mathrm{G}}(\omega) = \frac{1}{(\omega + i\eta)\hat{1} - \hat{\Omega}}. \tag{3.1}$$

Further, $\hat{\Omega} \in \mathcal{H}$ is a dynamical operator which governs the development of the studied physical system and $\mathcal{H}$ is the underlying vector space of operators and state vectors. In quantum mechanics, operator $\hat{\Omega}$ is the standard Hamiltonian $\hat{\mathrm{H}}$ and in classical physics it is a Lagrangean or Liouvillian, etc. Operator $\hat{\Omega}$ need not be Hermitean and, thus, we shall consider $\hat{\Omega}^\dagger \neq \hat{\Omega}$ throughout, so that generally the eigenvalues $\{\omega_k\}_{k=1}^K$ are complex numbers. The total number K of frequencies $\{\omega_k\}$ is any finite or infinite positive integer. The FPT determines this number exactly from the uniqueness conditions of the Padé polynomial quotient for the Green function, which is a power series with the given time signal points as the expansion coefficients. This is in sharp contrast to guessing K as customarily done in many other parametric estimators, e.g. LP, HLSVD, etc. Even the usual Hamiltonians are often converted to complex operators through the concept of non-Hermitean optical-type absorbing potentials that could mimic absorption of the incoming particle flux by the target. Customarily, a complex 'Hamiltonian' is constructed by introducing a purely imaginary absorbing potential $i\mathrm{W}$ according to the substitution $\hat{\Omega} \longrightarrow \hat{\Omega} + i\mathrm{W}\hat{1}$. In the interaction region, the value W is selected to be negligibly small and it is taken as being positive in the asymptotic region of scattering. The net effect of these so-called absorbing boundary conditions (ABC) [204]–[206] is to remove, i.e. to absorb the outgoing wave packet corresponding to direct scattering states lying in the asymptotic domain. The potential $i\mathrm{W}$ is otherwise artificial and rather arbitrary. What matters

is that iW does not change the physics at all. Rather, a flexible choice of W could help numerical computations. The introduction of complex 'Hamiltonians' is essential for treating resonances that are inherently different from genuine (physical) bound states. True square-integrable bound-state eigenvectors are associated with discrete negative eigenenergies and Hermitean Hamiltonians. Resonant states are also square-integrable, but they are linked to complex energies with positive real parts that belong to a continuum. Continuum states of Hermitean Hamiltonians are not normalizable and, therefore, do not represent physical states, so that they cannot describe particles. The concept of the ABC comes to rescue the situation via the introduction of complex 'Hamiltonians' with normalizable scattering wavefunctions as proper physical states. Note that complex 'Hamiltonians' could also follow from the Dyson device which is based upon the so-called adiabatic theorem from formal scattering theory [145]–[157]. This amounts to a modification consisting of including the damping factor $i\eta$ from $\hat{G}(\omega)$ directly into the total interaction potential $\hat{V} \longrightarrow \hat{V}e^{i\eta}$ which is a part of $\hat{\Omega}$. Alternatively, one could use the well-known rotated coordinate method (RCM) [207]–[210], which is also called the complex scaling method, to construct a complex 'Hamiltonian' $\hat{\Omega}$, which is non-Hermitean[1]. The crucial practical advantage of these circumstances that render operator $\hat{\Omega}$ complex is that its spectrum does not need to include explicitly continuum states that are known as difficult to handle in computations. Such states could be approximately represented by a pseudo-continuum, which is a collection of pure discrete states at complex energies encompassing both bound states and resonances. A pseudo-continuum or discretized continuum can be built from complex energies as a surrogate for a true continuum. For example, the behaviour of the Green function $\hat{G}(\omega)$ in the close vicinity of cuts or branch point singularities, can be described remarkably well by a sequence of poles of the FPT for $\hat{G}(\omega)$. This is essential, since no theory can pretend to describe scattering phenomena unless cuts and branch point singularities are treated in a satisfactory manner. An approximate description of a genuine continuum can also be achieved by the RCM in which a mapping from the real radial coordinate r to its complex counterpart $re^{i\theta}$ with a real scaling parameter θ leads to a non-Hermitean Hamiltonian $\hat{H}^{\dagger}(\theta) \neq \hat{H}(\theta)$. The full widths at half maxima of all the spectral peaks, that are embedded in a continuum to represent localized positive energy wave packets, determine the inverse lifetimes of every resonance in the spectrum of $\hat{\Omega}$. The infinitesimal number $\eta > 0$ secures regularity of $\hat{G}(\omega)$ for those values of ω that belong to the set of the eigenfrequencies $\{\omega_k\}$ of $\hat{\Omega}$. Once the calculation has been completed, the limit $\eta \to 0^+$ or $\eta \to 0^-$ should be taken depending upon whether the outgoing or incoming boundary conditions are imposed. The superscripts $\pm$ indicate that η should tend to zero through positive/negative numbers, respectively. In principle, we could omit η hereafter

[1] It has been often been stated that the RCM was introduced in 1971 by Baslev and Combes [210]. However, in reality, the RCM has been published at least three times before 1971, e.g. in 1960 by Zel'dovich [207], in 1961 by Dykhne and Chaplik [208] and in 1964 by Lovelace [209].

with the understanding that ω will stand for $\omega + i\eta$. However, this is unnecessary and we shall keep considering ω as being purely real, since $\hat{\Omega}$ will be conceived as a complex 'Hamiltonian'. In such a case, the eigenfrequencies ω_k of $\hat{\Omega}$ are complex valued, so that the resolvent $1/(\omega\hat{1} - \hat{\Omega}) = \sum_{k=1}^{K} \hat{\pi}_k/(\omega - \omega_k)$ is not singular, provided $\mathrm{Im}(\omega_k) \neq 0$, with $\hat{\pi}_k$ being the projection operator $\hat{\pi}_k = |\Upsilon_k)(\Upsilon_k|$ where $|\Upsilon_k)$ is the complete eigenfunction of the studied system obeying the stationary Schrödinger equation $\hat{\Omega}|\Upsilon_k) = \omega_k|\Upsilon_k)$. Clearly, if $\mathrm{Im}(\omega_k) = 0$, the Dyson damping $i\eta$ should be reintroduced.

A method which can provide an adequate spectral representation of the total Green operator or resolvent $\hat{\mathrm{G}}(\omega)$ would be one of the key inputs into an invaluable practical quantum-mechanical theory for scattering ($E \geq 0$) *and* spectroscopy ($E < 0$). If the operator $\hat{\mathrm{G}}(\omega)$ is available then all the observables for a collisional and a spectroscopic phenomenon could be obtained from the general Green function $\mathcal{G}_{if}(\omega) = (\Phi_{0f}|\hat{\mathrm{G}}(\omega)|\Phi_{0i})$. The diagonal elements are obtained for $|\Phi_{0i}) = |\Phi_{0f}) \equiv |\Phi_0)$ as $\mathcal{G}_{00}(\omega) \equiv \mathcal{G}(\omega)$

$$\mathcal{G}(\omega) \equiv (\Phi_0|\hat{\mathrm{G}}(\omega)|\Phi_0) \tag{3.2}$$

where $|\Phi_0)$ is the initial state of the system. It is possible to show that even the state vector $|\Phi_0)$ can be reconstructed from the known $\{C_n\}$ or $\{c_n\}$ [142]. Hereafter, we shall use the symmetric definition of the scalar product

$$(\zeta|\xi) = (\xi|\zeta) = \int \mathrm{d}V \zeta\xi \tag{3.3}$$

without conjugation of either of the two functions $(\zeta|$ and $|\xi)$. Here, $\mathrm{d}V$ is the differential of the volume element. The so-called local density of states $\rho_{00}(\omega) \equiv \rho(\omega)$ can be computed from the residues of $\mathcal{G}(\omega)$ at its singularities

$$\rho(\omega) = -\frac{1}{\pi} \lim_{\eta \to 0^+} \mathrm{Im}\{\mathcal{G}(\omega)\}. \tag{3.4}$$

An entirely generic system is presently subjected to our analysis by the general stationary and time-dependent methods of quantum mechanics. In the stationary methods, the complete set of eigenstates $\{|\Upsilon_k)\}$ of the Schrödinger operator $\hat{\Omega}$ will be available by obtaining the pair of the solutions $\{\omega_k, |\Upsilon_k)\}$ of the time-independent eigenvalue problem $\hat{\Omega}|\Upsilon_k) = \omega_k|\Upsilon_k)$ or by solving a more general eigenequation $f(\hat{\Omega})|\Upsilon_k) = f(\omega_k)|\Upsilon_k)$

$$\hat{\Omega}|\Upsilon_k) = \omega_k|\Upsilon_k) \qquad f(\hat{\Omega})|\Upsilon_k) = f(\omega_k)|\Upsilon_k) \tag{3.5}$$

where $1 \leq k \leq K$ and $f(\hat{\Omega})$ is any operator analytic function. *The main postulate of quantum mechanics is that the whole information of a general system under study is contained in the total wavefunctions* $\{|\Upsilon_k)\}$. This is known as completeness of the quantum-mechanical description of phenomena in nature. Such a circumstance is coherent with (3.5), since $\hat{\Omega}$ is assumed to carry the whole

information of the investigated object. This is formally expressed through the local closure relation for the exact orthonormalized basis $\{|\Upsilon_k)\}$

$$\sum_{k=1}^{K} \hat{\pi}_k = \hat{1} \qquad \hat{\pi}_k = |\Upsilon_k)(\Upsilon_k| \qquad (\Upsilon_{k'}|\Upsilon_k) = c_0 \delta_{k,k'} \tag{3.6}$$

where $\hat{\pi}_k$ is the projection operator and $\delta_{k,k'}$ is the Kronecker δ-symbol

$$\delta_{k,k'} = \delta_{kk'} = \begin{cases} 1 & k' = k \\ 0 & k' \neq k. \end{cases} \tag{3.7}$$

Notice that the local completeness is used in (3.6) by limiting the summation to the K terms only. Nevertheless, as already pointed out, the non-negative integer K need not be finite in the subsequent analysis. The norm $\|\Upsilon_k\|$, as the number whose square is defined by the scalar product of $|\Upsilon_k)$ with $(\Upsilon_k|$, is set to be equal to $c_0 \neq 0$

$$\|\Upsilon_k\|^2 \equiv (\Upsilon_k|\Upsilon_k) = c_0 \qquad (k = 1, 2, 3, \ldots). \tag{3.8}$$

This norm is not necessarily positive definite and it could be even complex for a complex c_0. The sum over k in (3.6) should, in principle, include integration over the continuum part of the spectrum of $\hat{\Omega}$. This is presently omitted, since resonances are taken into account through the spectrum of the non-Hermitean dynamic operator $\hat{\Omega}$. Inserting the representation (3.6) of the unit operator $\hat{1}$ into $\hat{G}(\omega) = \hat{G}(\omega)\hat{1}$ leads at once to the following spectral representation of the Green resolvent $\hat{G}(\omega)$ and the Green function $\mathcal{G}(\omega)$ from (3.2)

$$\hat{G}(\omega) = \sum_{k=1}^{K} \frac{\hat{\pi}_k}{\omega - \omega_k} \qquad \mathcal{G}(\omega) = \sum_{k=1}^{K} \frac{d_k}{\omega - \omega_k}. \tag{3.9}$$

Here, d_k are the complex amplitudes representing the residues associated with the eigenfrequencies $\{\omega_k\}$ that are the poles of the Green function $\mathcal{G}(\omega)$

$$d_k = (\Phi_0|\Upsilon_k)^2 \tag{3.10}$$

where (3.3) is employed with the property $(\Phi_0|\Upsilon_k) = (\Upsilon_k|\Phi_0)$. Let us temporarily assume that we are given the eigensolutions $\{\omega_k, |\Upsilon_k)\}$, but *not* the 'pseudo-Hamiltonian' $\hat{\Omega}$ itself. Then, $\hat{\Omega}$ is retrieved from its implicit definition

$$\hat{\Omega} = \sum_{k=1}^{K} \omega_k \hat{\pi}_k \qquad f(\hat{\Omega}) = \sum_{k=1}^{K} f(\omega_k)\hat{\pi}_k \tag{3.11}$$

where $f(\hat{\Omega})$ is any operator analytic function. In the particular case with $f(\hat{\Omega}) = (\omega\hat{1} - \hat{\Omega})^{-1} = \hat{G}(\omega)$, the result (3.9) follows again from (3.11). According to the Cauchy theorem, the local spectral representation of the Green operator $\hat{G}(\omega)$ is fully determined by the set $\{\omega_k, \hat{\pi}_k\}$ of its singularities $\{\omega_k\}$ and the corresponding operator residues $\{\hat{\pi}_k\}$.

Chapter 4

The time-dependent Schrödinger equation

A formalism which is equivalent to the preceding stationary treatment is provided by the time-dependent Schrödinger equation [143]–[157]

$$i\frac{\partial}{\partial t}|\Phi(t)) = \hat{\Omega}|\Phi(t)). \tag{4.1}$$

In the Schrödinger picture of quantum mechanics, operators are stationary and wavefunctions are time-dependent. The Schrödinger states $|\Upsilon)$ and $|\Phi)$ are interrelated by the usual one-sided Fourier integral with the semi-infinite limits [145, 157]

$$|\Upsilon(u)) = \frac{1}{2\pi}\int_0^\infty \mathrm{d}t\, \mathrm{e}^{i\omega t}\,|\Phi(t)) \qquad |\Upsilon_k) = \frac{1}{2\pi}\int_0^\infty \mathrm{d}t\, \mathrm{e}^{i\omega_k t}\,|\Phi(t)) \tag{4.2}$$

where $|\Upsilon_k) \equiv |\Upsilon(u_k))$. Here $u = \exp(-i\omega\tau)$ and $u_k = \exp(-i\omega_k\tau)$ are present through $\exp(i\omega t)$ and $\exp(i\omega_k t)$ written as $\exp(i\omega t) = u^{-t/\tau}$ and $\exp(i\omega_k t) = u_k^{-t/\tau}$, respectively. For the stationary ansatz $\hat{\Omega}$ we know that (4.1) possesses the solution of the type

$$|\Phi(t)) = \hat{\mathrm{U}}(t)|\Phi_0) \qquad |\Phi_0) \equiv |\Phi(0)) \tag{4.3}$$

where $\hat{\mathrm{U}}(t)$ is the dynamical evolution/relaxation linear operator of the system

$$\hat{\mathrm{U}}(t) = \mathrm{e}^{-i\hat{\Omega}t}. \tag{4.4}$$

In (4.3), $|\Phi_0)$ represents the initial unnormalized state of the studied system at the time $t = 0$

$$\|\Phi_0\|^2 \equiv (\Phi_0|\Phi_0) = C_0 = c_0 \neq 0 \tag{4.5}$$

where the constant c_0 from (3.8) is not necessarily unity, but rather it could be a complex number. These preliminaries imply that, if $\hat{\Omega}$ and $|\Phi_0)$ are known, then the determinism of quantum mechanics prescribes the exact knowledge of the

state $|\Phi(t))$ of the system at any later instant $t > 0$. The spectral representation of the operator $\hat{\mathrm{U}}(t)$ follows from using (3.11) where we set $f(\hat{\Omega}) = \mathrm{e}^{-i\hat{\Omega}t}$

$$\hat{\mathrm{U}}(t) = \sum_{k=1}^{K} \mathrm{e}^{-i\omega_k t}\hat{\pi}_k \qquad \mathrm{Im}(\omega_k) < 0. \tag{4.6}$$

The state $|\Phi(t))$ at the instant t is obtained by propagating the initial well-prepared wave packet $|\Phi_0)$ from $t = 0$ to t via $\hat{\mathrm{U}}(t)$. Given $|\Phi_0)$ at $t = 0$, there will be a non-zero probability amplitude to find the system in the state $|\Phi(t))$ at the later time $t > 0$, if the two wave packets have a non-vanishing overlap. This overlap is found quantum-mechanically by projecting $|\Phi(t))$ onto $(\Phi_0|$ by means of their scalar product

$$C(t) = (\Phi_0|\Phi(t)) = (\Phi_0|\hat{\mathrm{U}}(t)|\Phi_0) \tag{4.7}$$

where (4.3) is used. The quantity $C(t)$ is called the auto-correlation function, since it measures the degree of correlations between the states $|\Phi(t = 0))$ and $|\Phi(t \neq 0))$ under the influence of the dynamical operator $\hat{\Omega}$. It is the presence of the operator $\hat{\Omega}$ which makes $|\Phi(t))$ differ from $|\Phi_0)$, as is obvious from (4.3). By switching the dynamics off ($\hat{\Omega} = \hat{0}$), the system would be allowed to remain indefinitely in the initial state, so that $C(t) = (\Phi_0|\Phi_0) = c_0 \neq 0$ as in (4.5) at any time t. The initial state $|\Phi_0)$ is assumed to be a non-zero state vector $|\Phi_0) \neq |0)$, as is customary in quantum mechanics. Propagating the initial 'zero vector' $|\Phi_0) = |0)$ from $t = 0$ and onward would inevitably lead to the trivial Schrödinger state $|\Phi(t)) = |0)$ at any later time $t \neq 0$ and, therefore, this possibility is excluded as uninteresting. At two times t' and t where $t' < t$, the state vector $|\Phi(t'))$ can be viewed as a delayed 'copy' of $|\Phi(t))$. Thus the overlap between the states $|\Phi(t))$ and $|\Phi(t'))$ for $t' < t$, as per the inner product $(\Phi(t')|\Phi(t))$, represents a measure of correlation between the state and its delayed copy. One such example follows from taking the time t' all the way back to the initial moment $t' = 0$. This yields the auto-correlation function $C(t) = (\Phi(0)|\Phi(t))$.

At large times t, the auto-correlation function $C(t)$ could be numerically unreliable due to instabilities that stem from considerable oscillations of the overlap $(\Phi_0|\Phi(t))$ as t increases. This could cause a heavy corruption of the quantity $C(t)$ with computational noise, e.g. round-off errors, ill-conditioning, etc. In general, the Green function $\mathcal{G}(\omega)$ in (3.2) exhibits singularities (poles, cuts, etc) because of the presence of the resolvent operator $\hat{\mathrm{G}}(\omega)$ from (3.1). It is often claimed in the literature that $C(t)$ is free from such singularities. This is untrue. The reason being that the severe oscillations of $C(t)$ for large t act as disguised singularities entirely similar to those encountered more transparently in $\mathcal{G}(\omega)$ [211]. This is obvious from the fact that both functions $C(t)$ and $\mathcal{G}(\omega)$ are built from the same 'Hamiltonian' $\hat{\Omega}$ which is a generator of an infinitesimal unitary transformation described by the evolution operator $\hat{\mathrm{U}}(t)$ [157]. Moreover,

the limits $t \longrightarrow \pm\infty$ in $C(t)$ are strictly equivalent to $\eta \longrightarrow 0^{\mp}$ in $\hat{\mathrm{G}}(\omega)$ in accordance with the so-called Abel limit from the formal scattering theory [142]. As a matter of fact, if one does not encounter instabilities in $C(t)$ in producing a spectrum, this could only mean that the asymptotic region $t \longrightarrow \pm\infty$ has not been approximately reached, with the consequence that some of the longer-lived transients did not have sufficient time to decay [211].

The term 'transient' usually refers to a time developing phenomenon which dies out after a sufficiently long time lag has elapsed ($t \longrightarrow \infty$) [72]. Such are the envelopes of, e.g. experimentally encoded time signals as a linear combination of damped exponentials $\{\exp(-i\omega_k t)\}$ with constant amplitudes $\{d_k\}$ such that all complex frequencies $\{\omega_k\}$ must have the negative imaginary parts $\mathrm{Im}(\omega_k) < 0$. Strictly speaking, these are *stable* transients. A more general meaning of the term transient in sequence-to-sequence transformations has been encountered in quantum-mechanical signal processing [17, 18]. There, the term *unstable* or *secular* transients has been used following [72, 212] whenever referring to, e.g. a time signal $c(t)$ in which one or more fundamental frequencies ω_k are exponentially diverging ($\mathrm{Im}(\omega_k) > 0$). Any finite or infinite sum of harmonics $\{\exp(-i\omega_k t)\}$ would diverge if at least one of the complex fundamental frequencies is secular $\mathrm{Im}(\omega_k) > 0$. In such a case, this whole mentioned sum of exponentials $\{\exp(-i\omega_k t)\}$, i.e. the signal $c(t)$ would cease to have any direct physical meaning, since $|c(t)| \longrightarrow \infty$ as $t \longrightarrow \infty$. This situation will remain incurable as long as one keeps adding up directly the partial sums of the time signal, as a linear combination of exponentials $\{\exp(-i\omega_k t)\}$. However, such a divergent set of partial sums could still be computed with a finite result $|c(t)| < \infty$ by means of certain sequence-to-sequence nonlinear transformations that are capable of converting divergent into convergent sequences through the concept of the anti-limit [72]. One such transformation is the PA [65]–[92], which can also accelerate convergence of slowly converging multi-dimensional sequences and/or series [158], [213]–[216].

The above-introduced two concepts, the stationary and the time dependent representations, are interrelated by means of the exact one-sided Fourier integrals (*not* to be confused with the Fourier transforms)

$$\mathcal{F}(\omega) = \frac{1}{2\pi}\int_0^{\infty} \mathrm{d}t\, \mathrm{e}^{i\omega t} C(t) \qquad C(t) = \int_0^{\infty} \mathrm{d}\omega\, \mathrm{e}^{-i\omega t}\mathcal{F}(\omega). \tag{4.8}$$

Both (3.1) and (4.4) contain the same operator $\hat{\Omega}$ which is the source of the complete information about the system. This feature, together with the unitarity and linearity of the standard Fourier operator, guarantees that the information is preserved when passing from the time to the frequency domain. Inserting (4.6) into (4.7) yields the result

$$C(t) = \sum_{k=1}^{K} d_k \mathrm{e}^{-i\omega_k t} \tag{4.9}$$

where the residues $\{d_k\}$ are given by (3.10). By definition, the quantities $\{d_k\}$ measure the extent of the squared projection of the state $|\Upsilon_k)$ onto $|\Phi_0)$. Thus the amplitudes $\{d_k\}$ are the weights that carry information about the strength of the contributions of individual normal/natural mode frequencies $\{\omega_k\}$ to $C(t)$ in (4.9). The magnitudes $\{|d_k|\}$ are the intensities of the non-orthogonal 'harmonics' $\{\exp(-i\omega_k t)\}$ featuring as the principal components that constitute the auto-correlation function $C(t)$. Moreover, $\phi_k = \text{Arg}(d_k)$ is the phase of $C(t)$. Substituting $C(t)$ from (4.9) into (4.8) and carrying out the time integral exactly gives a *theoretically generated* complex spectrum $\mathcal{F}(\omega)$ as

$$\mathcal{F}(\omega) = -i \sum_{k=1}^{K} \frac{d_k}{\omega - \omega_k} \tag{4.10}$$

provided that $\text{Im}(\omega_k) < 0$. The result (4.10) for $i\mathcal{F}(\omega)$ agrees fully with the Green function $\mathcal{G}(\omega)$ from (3.9) as it should. The real quantities, e.g. the magnitude $|\mathcal{F}(\omega)|$, power $|\mathcal{F}(\omega)|^2$, absorption and dispersion spectra can all be obtained directly from $\mathcal{F}(\omega)$. Under ideal conditions of absence of noise and with no initial time delay, the absorption and dispersion spectra are given by $\text{Re}\{\mathcal{F}(\omega)\}$ and $\text{Im}\{\mathcal{F}(\omega)\}$, respectively. Of course, in practice, these conditions are not fulfilled and the absorption and dispersion spectra might differ from $\text{Re}\{\mathcal{F}(\omega)\}$ and $\text{Im}\{\mathcal{F}(\omega)\}$, respectively. In such cases certain alternative definitions of the absorption and dispersion spectra could be used [9, 115]. The resonance parameters from (4.10) are the position, width, height and phase of the kth peak given by $\text{Re}(\omega_k)$, $\text{Im}(\omega_k)$, $|d_k|$, $\text{Arg}(\omega_k)$, respectively. The auto-correlation function $C(t)$ from (4.7) is identified with an instantaneous transition probability amplitude for the passage of the system from $|\Phi(t = 0))$ to $|\Phi(t \neq 0))$. Then the survival probability amplitude for the state $|\Phi(t))$ can be obtained in the limit of $C(t)$ for large times $|t| \longrightarrow \infty$ [17]. Such a time limit is crucial for any collision problem in order to secure that the full scattering states are reduced to the appropriate *free* wave packets [157, 211]. In spectroscopy, the total acquisition time or epoch (T) of time-dependent observables should be sufficiently long to facilitate decays of all transient states, so that the physically relevant transitions could be unambiguously detected.

Chapter 5

Equivalence: auto-correlation functions and time signals

In practice, one equidistantly discretizes (digitizes) the continuous (analog) time variable t as $t = t_n = n\Delta t \equiv n\tau (n = 0, 1, 2, \ldots, N-1)$, where the non-negative integer n counts the time. The quantity τ is the time increment (the time lag) or the sampling time, which is also called the dwell time $\tau = T/N$ in ICR mass spectroscopy [7, 9]. As in chapter 2 we shall write

$$C_n \equiv C(n\tau) \qquad |\Phi_n) \equiv |\Phi(n\tau)) \tag{5.1}$$

for the discrete counterparts of $C(t)$ and $|\Phi(t))$, respectively. In practice, T is finite which implies that the time Fourier integrals from (4.8) should have the upper limits equal to T with $T < \infty$. In such a case, the Fourier integral $F(\omega)$ of the auto-correlation function $C(t)$ is introduced as

$$F(\omega) = \frac{1}{T}\int_0^T \mathrm{d}t\, C(t)\, \mathrm{e}^{i\omega t}. \tag{5.2}$$

This expression can be discretized if the integral in (5.2) is represented by its Riemann sum F_k evaluated at the Fourier grid $\omega = \tilde{\omega}_k$

$$F_k = \frac{1}{N}\sum_{n=0}^{N-1} C_n\, \mathrm{e}^{in\tilde{\omega}_k\tau} = \frac{1}{N}\sum_{n=0}^{N-1} C_n \mathrm{e}^{2i\pi nk/N} \qquad \tilde{\omega}_k \equiv \frac{2\pi k}{T} = \frac{2\pi k}{N\tau} \tag{5.3}$$

where $(0 \leq k \leq N-1)$ and $F_k \equiv F(\tilde{\omega}_k)$. This is the complex form of the discrete Fourier *transform* (DFT) [64], which is defined only at the Fourier frequencies $\omega = \tilde{\omega}_k$. If we multiply (5.3) by $(1/N)\exp(-im\tilde{\omega}_k\tau)$ and sum the result over k from 0 to $N-1$, we shall retrieve all the elements from the original set $\{C_n\}$ through the inverse discrete Fourier transform (IDFT)

$$C_n = \sum_{k=0}^{N-1} F_k \mathrm{e}^{-in\tilde{\omega}_k\tau} = \sum_{k=0}^{N-1} F_k \mathrm{e}^{-2i\pi nk/N} \qquad (0 \leq n \leq N-1) \tag{5.4}$$

where the orthogonalization property of the harmonic basis set functions $\{|\exp(-i\tilde{\omega}_k\tau))\}_{k=0}^{N-1}$ is employed [64]

$$\frac{1}{N}\sum_{k=0}^{N-1} e^{2i\pi(n-m)k/N} = \delta_{n,m} \tag{5.5}$$

where $\delta_{n,m}$ is the Kronecker δ-symbol from (3.7). The Fourier frequencies $\{\tilde{\omega}_k\}$ from (5.3) are purely real, so that the exponentials in (5.4) are unattenuated. Unlike (4.9), where the elements ω_k from the set $\{\omega_k\}$ are the unknown peak parameters, the frequencies $\tilde{\omega}_k$ from (5.3) are fixed in advance and this latter feature leads to linearity of both the DFT and the IDFT. The spectrum F_k^{bl}, which is equal to F_k within a fixed frequency interval/band and zero elsewhere, is called the band-limited (bl) Fourier transform [64, 113]. In (5.3) and (5.4) one encounters three sequences $\{C_n, F_k, |\exp(-i\tilde{\omega}_k\tau))\}_{n,k=0}^{N-1}$, each of which is of length N. A direct computation by means of the DFT would require N^2 multiplications that are drastically reduced to only $N\log_2 N$ multiplications in the fast Fourier transform (FFT) [176].

Here, it is pertinent to briefly recall several important efforts aimed at accomplishing the 'fast' $N^2 \longrightarrow N\log_2 N$ reduction. Certainly, significant computational costs required by N^2 multiplications were the main reason for the lack of widespread usage of the DFT up to 1965. Nevertheless, there was another reason which delayed computerized applications of the DFT, and that was a lack of awareness of certain of the most critical achievements in the Fourier analysis from the past. For example, in 1942 Danielson and Lanczos [46] showed in a transparent way, as recapitulated more recently in [53, 54, 176], that the N^2 computational complexity of the DFT can be drastically reduced to only $N\log_2 N$ multiplications, thus yielding the FFT, provided that the signal length N is a composite number, $N = 2^m (m = 0, 1, 2, 3, \ldots)$. Danielson and Lanczos [46] refer to the papers by Runge [44] from 1903 for the original source of their algorithm. In the period 1939–1963 there have been several other revivals of the $N^2 \longrightarrow N\log_2 N$ reduction with the ensuing transformation of the DFT to the FFT (Stumpff [45], Good [47], Thomas [48], etc). Despite this vigorous development, which originally started by Gauss in 1805 [43], and published in his collected works, it was not until 1965 that the FFT became widely known through its reinvention by Cooley and Tukey [49] who were apparently unaware of any of the previous related works from [43]–[48]. However, soon after publication of the paper by Cooley and Tukey [49], an important comment was made in 1966 by Rudnick [50] who pointed out that the $N^2 \longrightarrow N\log_2 N$ computational saving in the FFT had been achieved already in 1942 by Danielson and Lanczos [46]. Following Rudnick [50], in 1967 Cooley *et al* [51] attempted to make a chronological overview of the introductions of the FFT prior to Cooley and Tukey [49]. They stated that [49] was more general in that it considered other alternatives to $N = 2^m$ (e.g. N being a prime number), while still preserving the crucial $N\log_2 N$ expediency. However, subsequently, it has been categorically

recommended, in e.g. *Numerical Recipes* [176], that one should use the FFT *only* with the signal length N being a power of two, in which case the algorithm from the work of Cooley and Tukey [49] coincides with that of Danielson and Lanczos [46]. It has been further pointed out in [176] that when N is not a power of two one should do either zero padding to the nearest power of two or use the LP method to predict the missing time signal points and thus achieve the needed length $N = 2^m$. Although Cooley and Tukey [49] were not the first to invent the FFT, they were the first in the modern computer era to succeed in reviving an unprecedented interest in the FFT, which after 1965 indeed revolutionized many sciences and technologies yielding profound benefits, especially to MR physics with most prominent applications in chemistry, biology and medicine. This comes as no surprise, since a computation by means of the DFT (N^2) for, e.g. $N = 10^6$ which might take even two weeks could be done within 30 seconds of the CPU (Central Processing Unit) through the use of the FFT ($N\log_2 N$) [176].

Due to the exponential nature of the evolution operator (4.4), construction of its discrete counterpart $\hat{\mathrm{U}}(t) = \hat{\mathrm{U}}(t_n) = \hat{\mathrm{U}}(n\tau)$ at the time $t = t_n \equiv n\tau$ is done simply through raising the ansatz $\hat{\mathrm{U}}(\tau)$ to the nth power

$$\hat{\mathrm{U}}(n\tau) = \hat{\mathrm{U}}^n(\tau) \qquad \hat{\mathrm{U}}(\tau) = \mathrm{e}^{-i\hat{\Omega}\tau} \Longrightarrow |\Phi_n) = \hat{\mathrm{U}}^n(\tau)|\Phi_0). \tag{5.6}$$

The set $\{|\Phi_n)\}$ represents the Schrödinger basis which could be used for diagonalization of the evolution/relaxation matrix $\mathbf{U}$. In linear programming and engineering literature [217]–[220], the set $\{|\Phi_n)\}$ is called the Krylov basis [221]. The same name is also used in quantum chemistry [108]–[116]. In quantum-mechanical signal processing, the term Schrödinger basis is more transparent, since it points directly at the quantum-mechanical origin of the state functions $\{|\Phi_n)\}$ that *do stem from the Schrödinger equation* (4.1). Nevertheless, to avoid potential confusion across interdisciplinary fields, one should always bear in mind that the Schrödinger and Krylov basis are two different nomenclatures for the same set $\{|\Phi_n)\}$ and, for this reason, we shall interchangeably use both terminologies. The discrete/digital counterpart of (4.6) can be written as

$$\hat{\mathrm{U}}^n(\tau) = \sum_{k=1}^{K} \mathrm{e}^{-in\omega_k\tau}\hat{\pi}_k \qquad \mathrm{Im}(\omega_k) < 0. \tag{5.7}$$

For any operator analytic function $f(\hat{\mathrm{U}})$, the eigenproblems follow as in (3.5)

$$\hat{\mathrm{U}}(\tau)|\Upsilon_k) = u_k|\Upsilon_k) \qquad u_k = \mathrm{e}^{-i\omega_k\tau} \qquad f(\hat{\mathrm{U}})|\Upsilon_k) = f(u_k)|\Upsilon_k). \tag{5.8}$$

Use of (4.7), (4.9) and (5.6) gives the discrete auto-correlation function

$$C_n = (\Phi_0|\Phi_n) = (\Phi_0|\hat{\mathrm{U}}^n(\tau)|\Phi_0) \qquad C_n = \sum_{k=1}^{K} d_k u_k^n. \tag{5.9}$$