On Fer and Floquet-Magnus Expansions: Applications in Solid-State Nuclear Magnetic Resonance and Physics

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OUTLINE

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- Average Hamiltonian Theory

C. **Alternative Expansion Approaches Used Methods in SS-NMR**

- Fer Expansion
- Floquet-Magnus Expansion

D. Applications of Fer and Floquet-Magnus expansion in SS-SNMR

E. **Applications of Fer and Floquet-Magnus expansion in Physics**

A. Background of NMR: Solid-State NMR

- **NMR is an extraordinary versatile technique which started in Physics In 1945 and has spread with great success to Chemistry, Biochemistry, Biology, and Medicine, finding applications also in Geophysics, Archeology, Pharmacy, etc...**

- **Hardly any discipline has remained untouched by NMR.**

- **It is practiced in scientific labs everywhere, and no doubt before long will be found on the moon.**

- **NMR has proved useful in elucidating problems in all forms of matter. In this talk we consider applications of NMR to solid state: Solid-State NMR**

BRIEF HISTORY OF NMR

- **1920's** **Physicists Have Great Success With Quantum Theory**
- **1921** **Stern and Gerlach Carry out Atomic and Molecular Beam Experiments**
- **1925/27** **Schrödinger/ Heisenberg/ Dirac Formulate The New Quantum Mechanics**
- **1936** **Gorter Attempts Experiments Using The Resonance Property of Nuclear Spin**
- 1937 Rabi Predicts and Observes Nuclear Magnetic Resonance
- 1944 Rabi awarded the Nobel prize for physics

Rabi was given this prize for his work on 'molecular beams, esp reson

Isidor Isaac Rabi

- Normally, credit for NMR first observation should go to Rabi and co-workers (1939) who used a beam of silver atoms
- The noticeable change in the fluxes of beams representing the different energy states of the nuclear magnetic moments was the detection of transitions

Ernest Lawrence (left), Enrico Fermi (center), and Isidor Rabi (right)

However, the term NMR has come to be used as a convention for experiments, which differ from those of Rabi.

- The experiments set by the convention in respect of NMR are those through the detection of the transitions with the energy absorbed from the RF field rather than through changes in the particle flux reaching a detector as in the beam experiments.

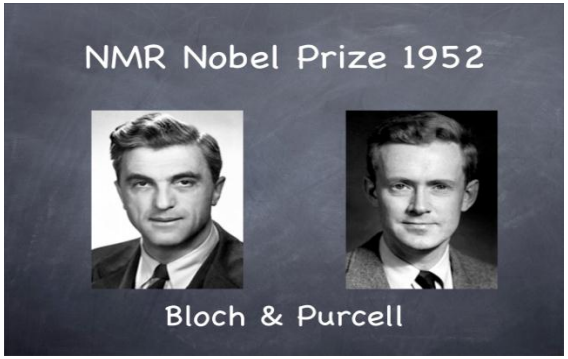
- Next, the term NMR is commonly reserved for phenomena occurring in bulk matter rather than in a beam of essentially non-interacting atoms.

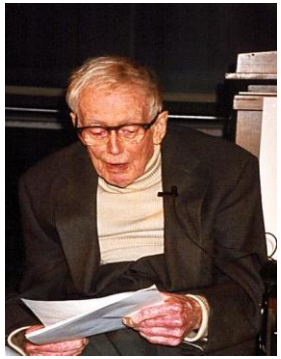
1945 Purcell, Torey and Pound observe NMR in a bulk material: paraffin (30 MHz)

1945 Bloch, Hansen and Packard observe NMR in solution: H_2O (8 MHz)

1952 Bloch and Purcell Share The Nobel Prize in Physics

This prize was awarded "for their development of new methods for nuclear magnetic precision Measurement and discoveries in connection therewith"





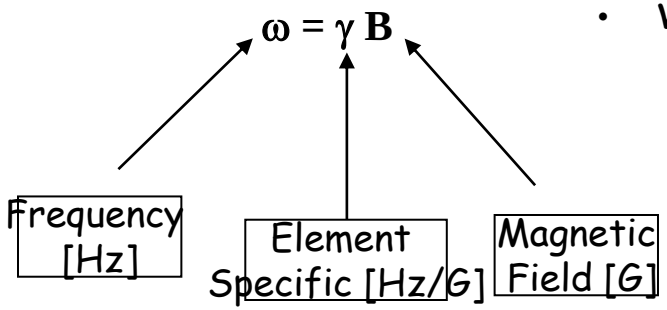
Edwards Purcell Dec. 10, 1995 Golden Jubilee of NMR – Harvard -

DEFINITION OF NMR

- When an atomic nucleus is placed in a magnetic field, the ground state will split into different energy levels proportional to the strength of the magnetic field. This effect is known as Zeeman Splitting.
- A constant magnetic field breaks the degeneracy of the energy levels of an atomic nucleus with spin. If the Nuclear spin is I, then 2I + 1 sub-levels appear.

NMR: BRANCH OF SPECTROSCOPY

Nuclear magnetic moments precess like "tops" in a magnetic field with a frequency



- When a time-dependent RF electromagnetic field of appropriate frequency is applied, energy can be absorbed by certain nuclei, which are consequently promoted to higher levels. This is the physical phenomenon of NMR.
- While the Zeeman interaction is useful for identifying different types of nuclei placed in magnetic fields, structural and dynamic information may be obtained by considering other magnetic and electronic interactions coupling with the nucleus.

Nuclear magnetic moments of a sample will precess at different frequencies if the field has a spatial dependence $B(z) \rightarrow \omega(z)$.

- A spinning charge generates a magnetic field, as shown by the animation on the right.
- The resulting spin-magnet has a magnetic moment (μ) proportional to the spin.

B= 9.6 x 10^4 G	ω [MHz]	γ [Hz/G]
^{1}H	400	4,250
^{19}F	376	3,995
^{15}N	40.56	431

- **Nuclear spin dynamics constitutes the basis for NMR, which is a very powerful spectroscopy technique that exploits the interaction between nuclear spins and magnetic fields.**

$$H = \sum_{\Lambda} H^{\Lambda}$$

$$H(t) = H_Z + H_{RF}(t) + H_{CS}(t) + H_J(t) + H_D(t) + H_Q(t)$$

Relative Strength of NMR Interactions

Interaction	Chemical Shift	J-Coupling	Dipolar	Quadrupolar
Liquids	10^3 -10^4 [Hz]	10 - 10^2 [Hz]	0 [Hz]	0 [Hz]
Solids	10^3 – 10^4 [Hz]	10 – 10^2 [Hz]	2 x 10^4 [Hz]	10^5 - 10^6 [Hz]

$$H_Z = -\sum_i \gamma_{[i]} \mathbf{B}^i \cdot \mathbf{I}_i = \sum_i \mathbf{n}_{B_0} \cdot \omega_{\mathrm{ref}}^{[i]} (1 + \boldsymbol{\delta}^i) \cdot \mathbf{I}_i,$$

$$H_{RF} = \sum_i |\omega_{RF}^i(t)|(I_{ix} \cos \phi_i + I_{iy} \sin \phi_i)$$

$$H_{CS} = \sum_i \omega_{CS,0}^i(t) I_{iz},$$

$$H_J = \sum_{ij} -\omega_{J_{iso},0}^{ij}(t) \frac{1}{\sqrt{3}} I_i.I_j + \omega_{J_{aniso},0}^{ij}(t) \frac{1}{\sqrt{6}} (3I_{iz}I_{jz} - I_i.I_j),$$

$$H_D = \sum_{i,j} \omega_{D,0}^{ij}(t) \frac{1}{\sqrt{6}} (3I_{iz}I_{jz} - I_i.I_j),$$

$$H_Q = \sum_i \omega_{Q,0}^i(t) \frac{1}{\sqrt{6}} (3I_{iz}^2 - I_i^2) + \frac{1}{2\omega_0^i} \Big\{ \omega_{Q,-2}^i(t) \omega_{Q,2}^i(t) (2I_i^2 - 2I_{iz}^2 - 1) I_{iz}) + \omega_{Q,-1}^i(t) \omega_{Q,1}^i(t) (4I_i^2 - 8I_{iz}^2 - 1) I_{iz} \Big\},$$

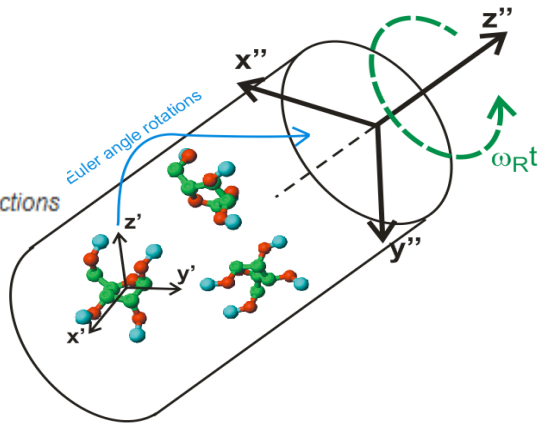
- **These interactions are perturbations to the Zeeman interaction**

- **SS-NMR experiments are subjected to various time-dependent perturbations of different frequencies, such as RF irradiations and MAS.**

- **The complexity of experiments due to the presence of the anisotropic interactions has made the use of mathematical methods necessary in solid-state NMR.**

Interaction	Magnitude (Hz)
Zeeman	10^8
Quadrupolar	10^6
Chemical Shift	10^3
Dipole	10^3
J	10

1. Magnitude of different NMR interactions



Discussion

Compare methods of solving Protein Structure

NMR	X-ray Crystallography
No crystal needed	Crystal
Can be used in solution	Solid only
Not good for large proteins, smaller molecules are comparable to X-ray	Generally higher solution
Can measure dynamics	Stationary
In vivo possible (imaging)	*In vitro*

Sensitivity Enhancement in NMR

Technique	*Authors*	*Enhancement (for ^{15}N)*
Fourier Transform NMR	Ernst and Anderson	~10-100
Polarization Transfer CP and INEPT	Hartmann & Hahn; Pines, Gibby & Waugh; Morris & Freeman	10
Indirect Detection (HSQC)	Bodenhausen and Ruben	~30
B_0 --200 to 800 MHz	Oxford, Bruker, Magnex	8
Cryoprobes	Peter Styles	2-4
TROSY	Pervushin, et.al.	2-5
High Frequency Dynamic Nuclear Polarization	Becerra,Gerfen,Prisner, McDermott, Un,Hall,Farrar, Rosay, Weis, Bennati, Hu, Bajaj, and RG[2]	**~ 50-400**

Nobel Prize

Wolf Prize x 2

Nobel Prize

$\varepsilon = 50\text{-}400$

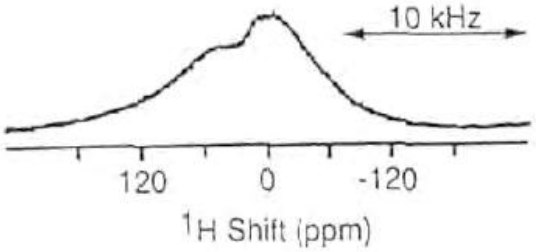
Significant consequences for NMR -- savings of ~ 2500 - 160,000 in time -- NEW SCIENCE !

From Robert Griffin, NMR Winter School: 01/2008

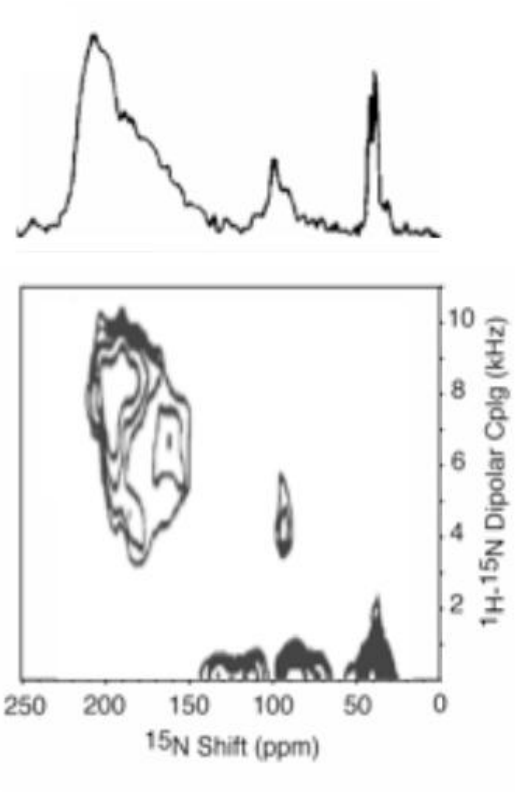
High Resolution

Improvements in solid-state NMR spectra of proteins.

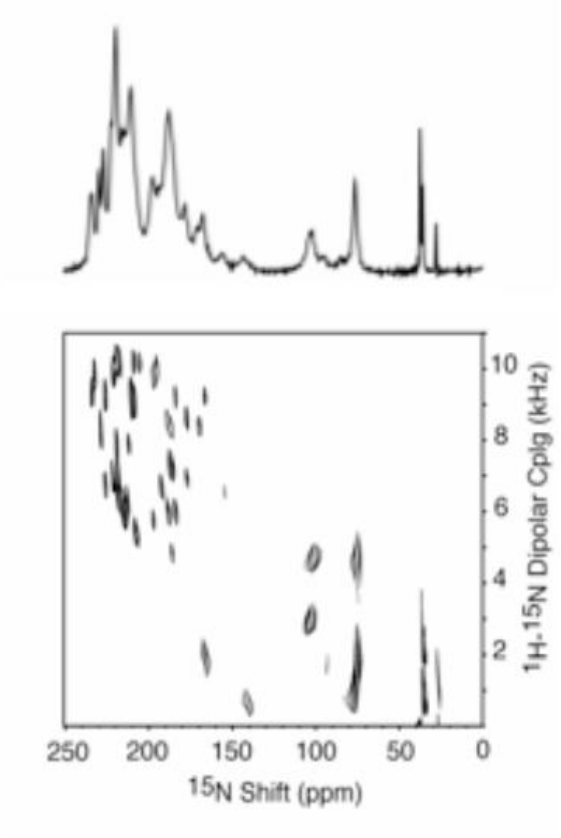
1976



1983

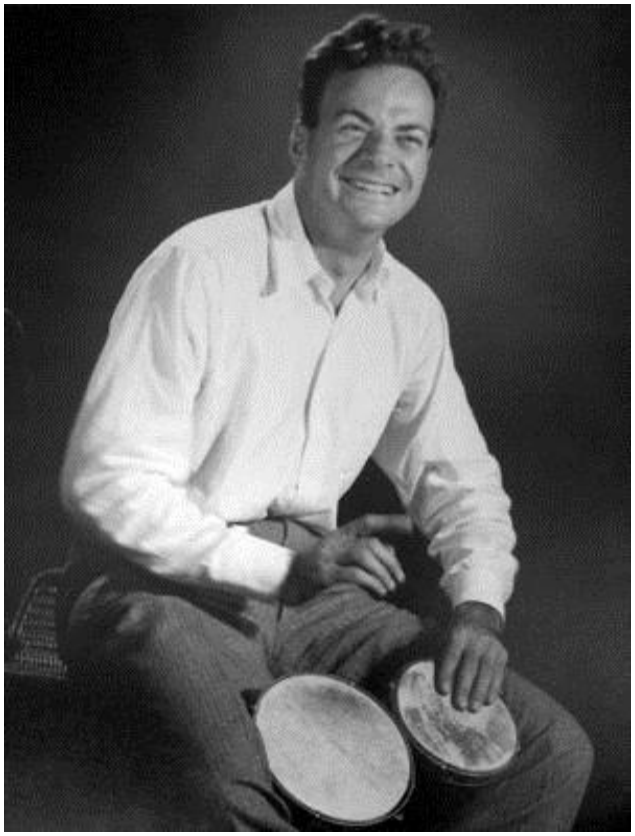


2004



From S. J. Opella, NMR Winter School: 01/ 2008

Excerpt from Richard Feynman's famous 1959 lecture "There's plenty of room at the bottom"



" ...we have friends in other fields---in biology, for instance. We physicists often look at them and say, You know the reason you fellows are making so little progress...you should use more mathematics, like we do.

They could answer us---but they're polite, so I'll answer for them: What you should do in order for us to make more rapid progress is to make the electron microscope 100 times better."

B. Commonly Used Methods in Solid-State NMR

- **Average Hamiltonian Theory**
- **Floquet theory**

The Schrödinger equation

The Schrödinger eqn describes the evolution of a quantum state. It is expressed in terms of bra and ket as:

$$\frac{d\,|\psi\rangle}{dt} = -i\,\hat{\mathcal{H}}\,|\psi\rangle$$

$$\frac{d\,\langle\psi|}{dt} = (-i\,\hat{\mathcal{H}}\,|\psi\rangle)^{\dagger} = i\,\langle\psi|\,\hat{\mathcal{H}}$$

In NMR we usually describe the state of the ensemble of spin states via the density operator, so we need to move from $|\psi\rangle$ to

$$|\psi\rangle\,\langle\psi|$$

Liouville-von Neumann equation

Take the time derivative of $|\psi\rangle\,\langle\psi|$ to relate the S.e., which applies to the wave function, to an equation applicable to $\hat{\rho}$:

$$\begin{aligned}\frac{d}{dt}(|\psi\rangle\,\langle\psi|) &= \left(\frac{d}{dt}|\psi\rangle\right)\langle\psi| + |\psi\rangle\left(\frac{d}{dt}\langle\psi|\right)\\ &= -i\,\hat{\mathcal{H}}\,|\psi\rangle\,\langle\psi| + i\,|\psi\rangle\,\langle\psi|\,\hat{\mathcal{H}}\\ &= i\left[|\psi\rangle\,\langle\psi|\,,\hat{\mathcal{H}}\right]\end{aligned}$$

Taking the ensemble average and assuming $\mathcal{H}$ to be identical over the ensemble, we get the Liouville-von Neumann equation:

$$\frac{d\hat{\rho}}{dt} = i\,\left[\hat{\rho},\hat{\mathcal{H}}\right] = -i\,\left[\hat{\mathcal{H}},\hat{\rho}\right]$$

L.v.N equation in terms of propagators

Start from the S.eq.

$$\frac{d}{dt}\,|\psi(t)\rangle = -i\,\hat{\mathcal{H}}(t)\,|\psi(t)\rangle$$

This becomes

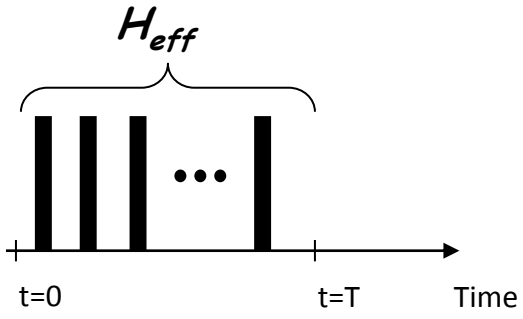
$$\frac{d}{dt}\hat{U}(t,t_0)\psi(t_0) = = -i\,\hat{\mathcal{H}}(t)\hat{U}(t,t_0)\,|\psi(t_0)\rangle$$

which holds for any initial $|\psi(0)\rangle$, hence we can write the L.v.N. equation directly in terms of the propagator:

$$\frac{d}{dt}\hat{U}(t,t_0) = = -i\,\hat{\mathcal{H}}(t)\hat{U}(t,t_0)$$

Average Hamiltonian Theory

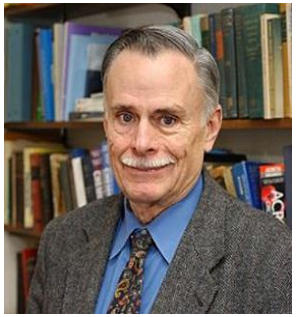
Basic Idea:



Replace sequence of RF perturbations and free evolution periods by and effective Hamiltonian

$$\psi(T) = e^{-iH_{eff}T}\,\psi(0)$$

- H_{eff} transform the initial state to the final state in time T.
- When H_{eff} = 0, spin at time T = spin at time 0.
- Note: state at t = 0 and t = T are identical , but not in between, where the state of the system may be evolving in a complicate way.


John Waugh (1929-2014)

Fundamental equation of AHT:

- U_{RF} represents the interaction associated with the sequence of RF pulses applied over a time t_c
- H_{int} refers to the system's internal Hamiltonian

LVN $$\rho(t_c) = U_{RF}U_{int}\rho(0)U_{int}^{-1}U_{RF}^{-1}$$

$$U_{RF} = \ldots U_3U_2U_1U_0$$

$$U_{int}(t,0) = \exp[-it_c(\overline{H}^0_{int} + \overline{H}^1_{int} + \cdots)]$$

ME is used in NMR spectroscopy with the AHT which is built up on the basis of the Conventional ME

$$\overline{H}^0_{int} = \frac{1}{t_c}\int_0^{t_c}\widetilde{H}_{int}(\tau)\mathrm{d}t$$

$$\overline{H}^1_{int} = \frac{-i}{2t_c}\int_0^{t_c}[\widetilde{H}_{int}(\tau), \int_0^{\tau}\widetilde{H}_{int}(\phi)\mathrm{d}\phi]\mathrm{d}\tau$$

$$\overline{H}^2_{int} = \ldots$$

Magnus Expansion

- **Wilhelm Magnus (1907-1990) made important contributions to a wide variety of fields in mathematics and mathematical physics. Among them, one of his long-lasting constructions: the so-called Magnus expansion (ME).**

$$i\frac{dU}{dt} = H(t)U(t)$$

$$\Longrightarrow$$

$$U(t) = e^{-i\Omega(t)}$$

- **ME was introduced as a tool to solve non-autonomous linear diff. Eq. for linear operators. In his seminal paper (1954), Magnus recognizes that his work was stimulated by results of Friedrichs (1953) on the mathematical aspects of the quantum theory of fields.**

With the help of the Wilcox Formula (1967)

$$i\frac{dU}{dt} = i\frac{d}{dt}\{e^{-i\Omega(t)}\} = \left\{\int_0^1 e^{-is\Omega(t)}\frac{d\Omega}{dt}e^{+is\Omega(t)ds}\right\}e^{-i\Omega(t)}$$

Introducing Liouville Operator

$$ad_\Omega Y = [\Omega, Y]$$

@ Basis of Magnus Expansion

$$\frac{d\Omega}{dt} = \phi^{-1}(-iad_\Omega)H(t)$$

$$\phi^{-1}(x) = \frac{x}{e^x - 1} = \sum_k \frac{B_k x^k}{k!}$$

where B_k are the Bernoulli numbers ($B_0 = 1$, $B_1 = -1/2$,

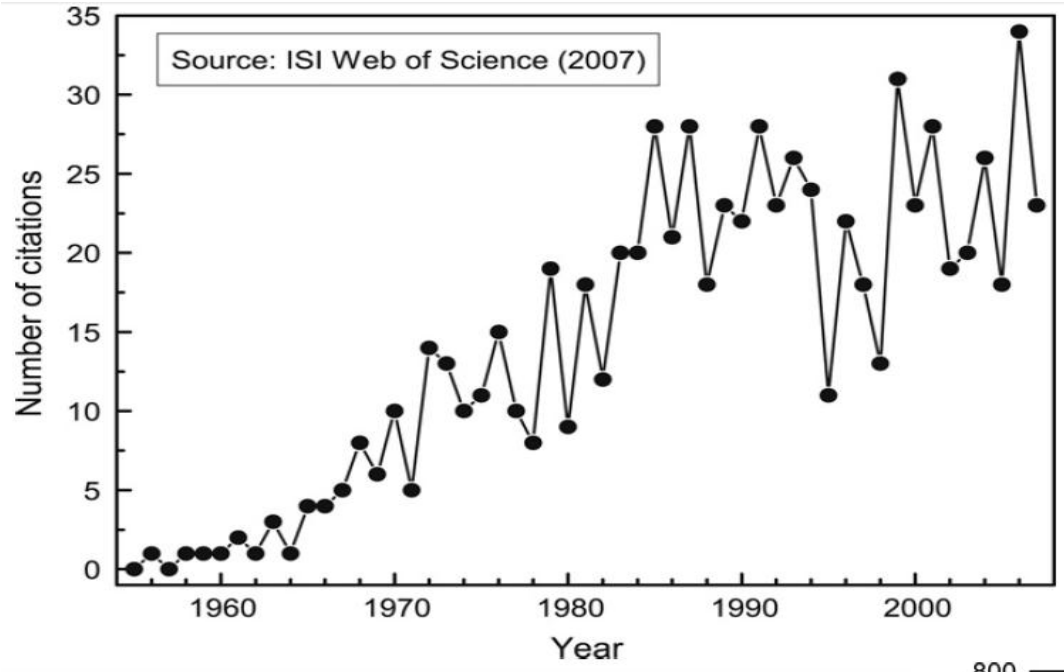
After substitution, the following **ME** is obtained

$$\frac{d\Omega}{dt} = \sum_k \frac{B_k}{k!}(-i)^k ad_\Omega^k \{H(t)\}$$

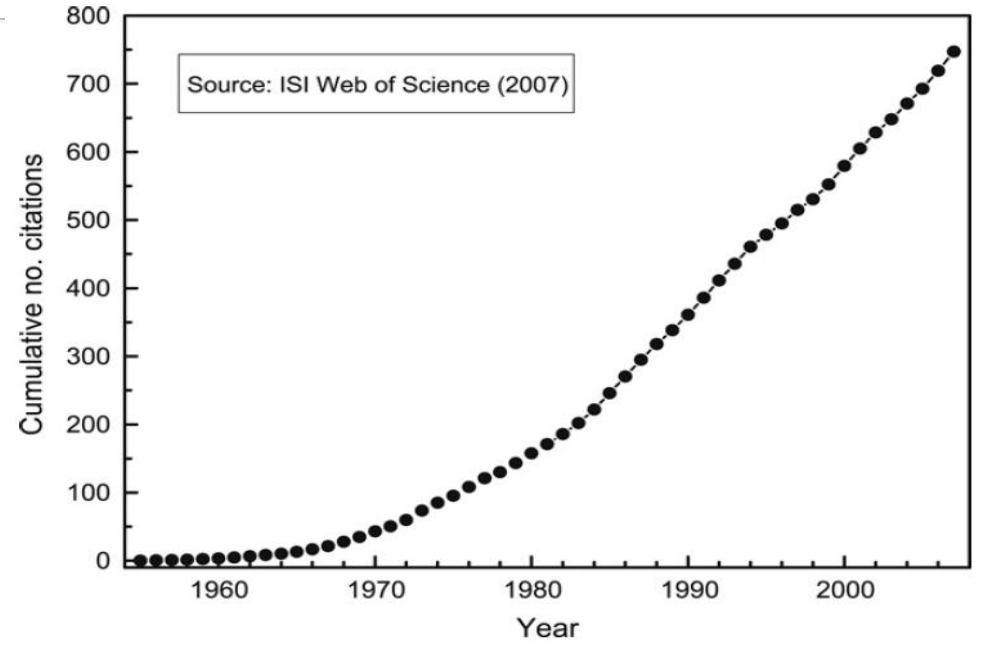
This equation justifies the name of **Exponential Perturbation**

Eugene Mananga and Thibault Charpentier, JCP, 135 (2011) 044109

Persistency of Magnus' Original Paper: Number of Citations per Year



Persistency of Magnus' Original Paper: Cumulative Number of Citations



S. Blane, F. Casas, J. A. Oteo, J. Ros, Physics Reports, 470 (2009) 151-238

Physical Applications of Magnus Expansion

ME has a strong bearing on both classical and quantum mechanics. Over the years, the ME has been one of the preferred options to deal with SE which, under different appearances, pervades the entire field of physics.

- **Nuclear magnetic resonance (Average Hamiltonian Theory):** Chuang (2004) envisaged the use of ME through NMR for the new field of quantum information processing and computing.

- **Nuclear, atomic and molecular physics**:

  (1) **the first physical application of ME dates back to 1963 by Robinson who published a new formalism to investigate multiple Coulomb excitations of deformed nuclei. The coulomb excitation process yields information about the low lying nuclear states.**

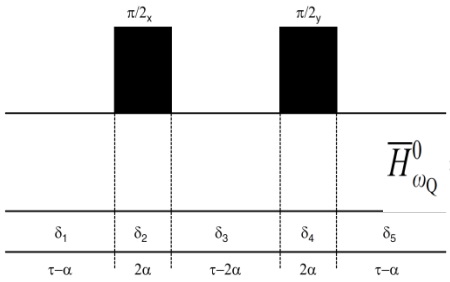
  (2) **in 1977, Eichler used ME to derive the transition amplitude and the cross section for K-shell ionization of atoms by heavy-ion impact. The use of ME allowed one to extend the studies to the ionization of light target atoms by much heavier projectile ions.**

- **Quantum field theory (QFT) and high energy physics:** The starting point of any QFT calculation is SE which is conventionally treated by time-dependent perturbation theory. So, the first question which arises is the connection between ME and Dyson-type series.
- **General Relativity:** Recent work of Miguel (2007) in numerical determination of time transfer in general relativity used also **ME**.
- **Geometric control of mechanical systems: ME** has been used in non-holonomic motion planning of systems without drift.

S. Blane, F. Casas, J. A. Oteo, J. Ros, Physics Reports, 470 (2009) 151-238

TWO SIMPLE APPLICATIONS OF AHT

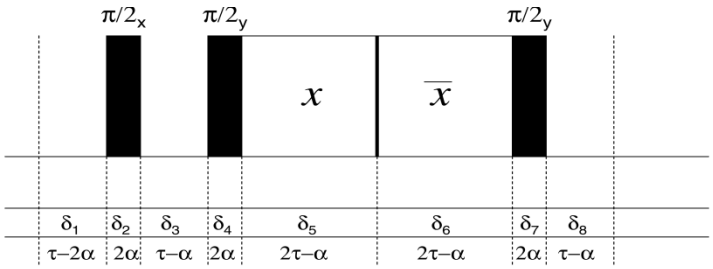
1.Solid Echo



$$\overline{H}^0_{\omega_Q} = \frac{4\alpha\omega_Q}{\pi\tau}[I_{x,2} - I_{z,2}] \qquad [22]$$

$$\overline{H}^0_{\omega_Q} = \frac{1}{7\tau}\frac{12\alpha\omega_Q}{\pi}I_{x,2} + \frac{4(3\alpha-\tau)}{7\tau}\omega_Q[I_{x,1}I_{x,1} + I_{y,1}I_{y,1} - 2I_{z,1}I_{z,1}] \qquad [23]$$

2. Magic Echo



Why Magic Echo performed better than Conventional Solid Echo?

Consider the convergence of the Magnus expansion

$$\overline{H}^1_{int}(ME) = \frac{1}{7\tau}\frac{\omega_Q^2}{\omega_{RF}^2}[\frac{18\sqrt{2}}{8}I_{y,1} - \frac{9}{16}(4\sqrt{2} + (-2+\sqrt{2}))I_{x,1}] \qquad [24]$$

$$\overline{H}^1_{int}(SE) = \frac{B}{3\tau}I_{y,1} - \frac{A}{3\tau}I_{z,1} + \frac{A}{3\tau}I_{x,1} \qquad [25]$$

$2\alpha = 2.0\mu s$

$\omega_Q = 125\text{KHz}$

$\tau = 100\mu s$

$\tau = 300\mu s$

$$A = \frac{18\alpha^2\omega_Q^2[\pi(-2+\sin(\frac{\pi\tau}{2\alpha})) + \sin(\frac{\pi\tau}{\alpha})]}{\pi^2} \qquad [26]$$

$$B = \frac{18\alpha^2\omega_Q^2[1+\cos(\frac{\pi\tau}{\alpha})]}{\pi^2} \qquad [18]$$

$$\overline{H}^1_{int}(ME) = 2.85I_{x,1} + 194.60I_{y,1} \qquad [27]$$

[28]

$$\overline{H}^1_{int}(SE) = -457.91I_{x,1} + 13.46I_{y,1} + 457.91I_{z,1}$$

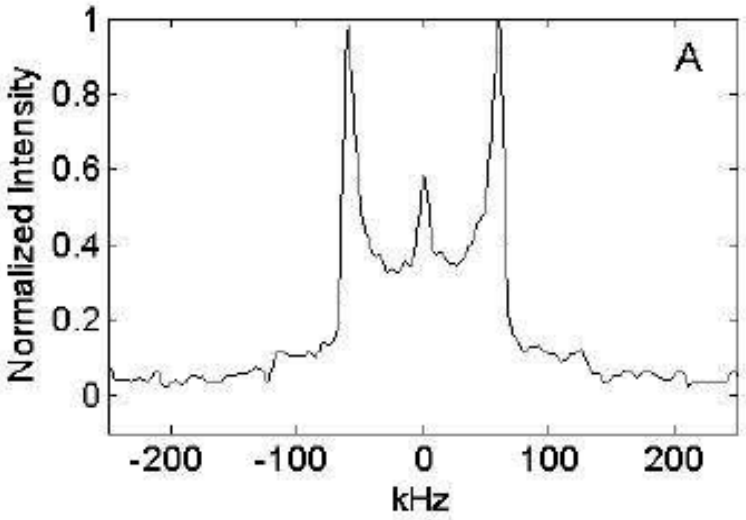
E. S. Mananga and al., JMR (2006) 296-303

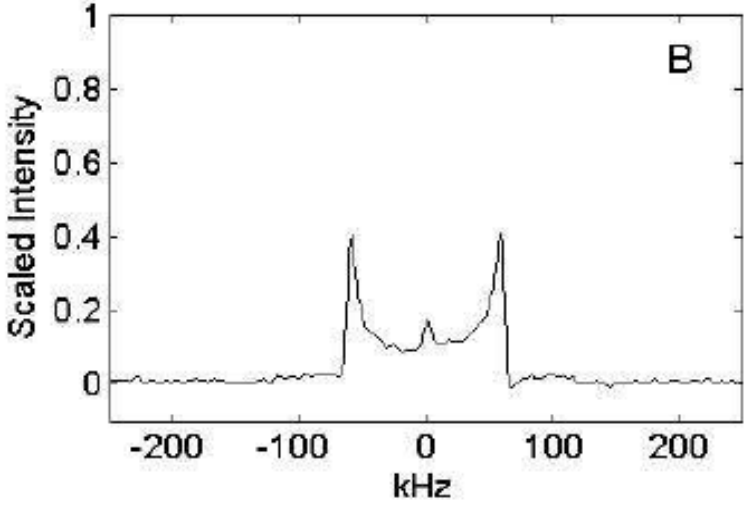
EXPERIMENTAL RESULTS

Magic Echo

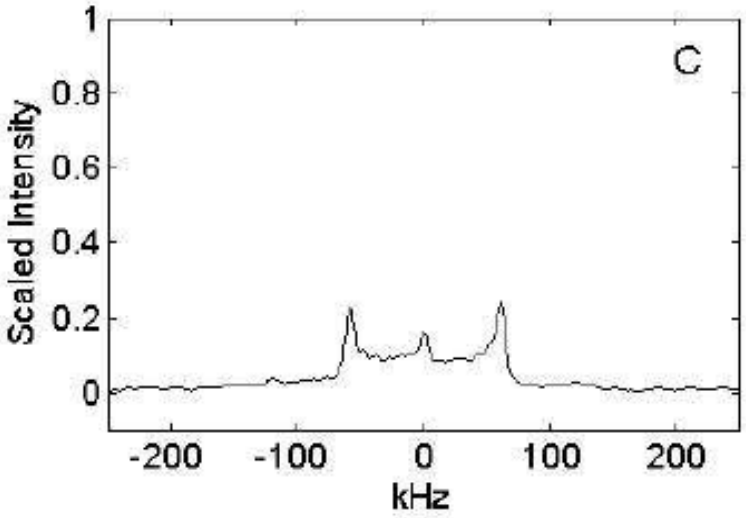
Solid Echo

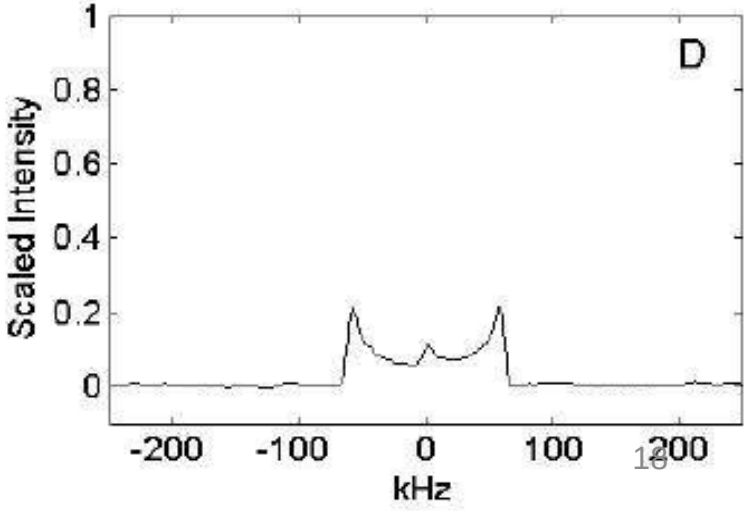
1.3 μs π/2 pulses





1.8 μs π/2 pulses

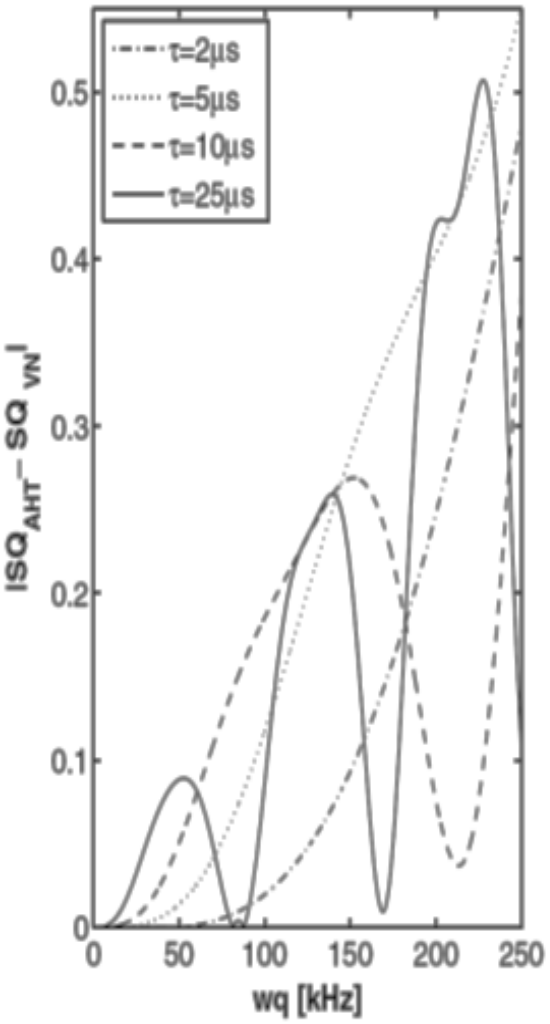




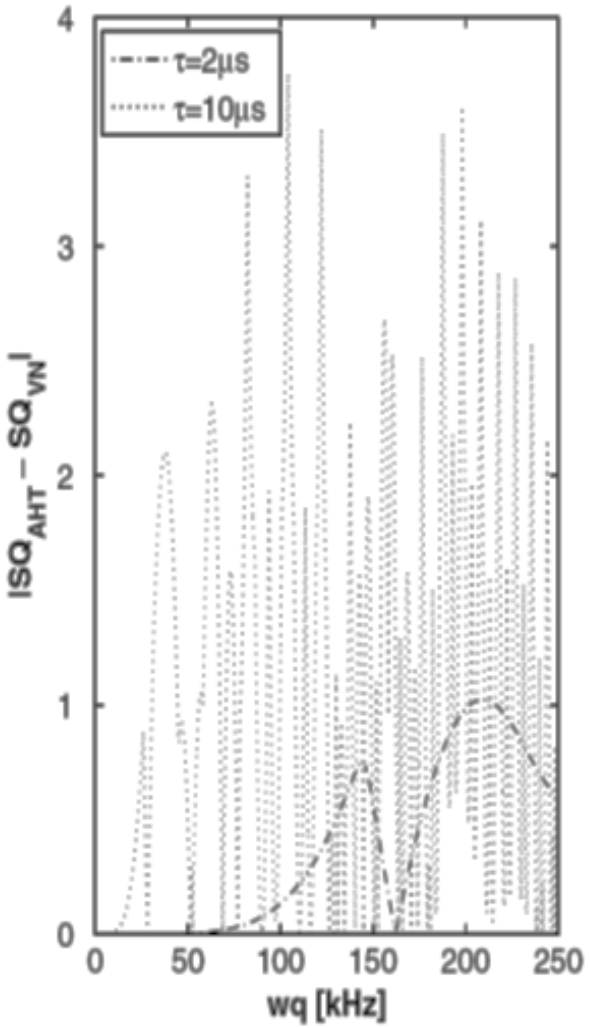
E. S. Mananga and al., JMR 185 (2007) 28-37

Probing the Validity of AHT for Spins: I=1, 3/2 & 5/2

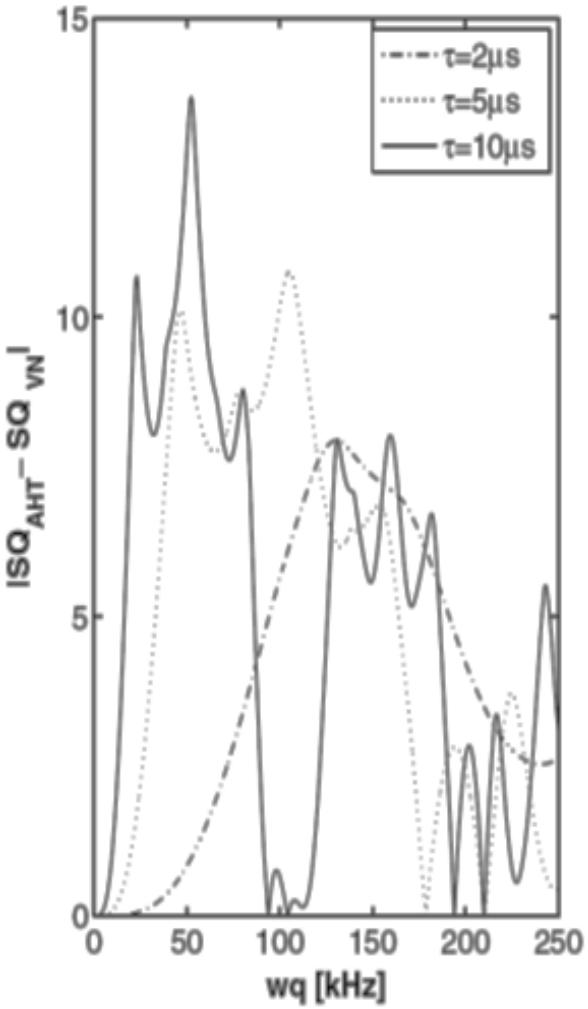
I = 1



I = 3/2



I = 5/2



E. S. Mananga and al., JMR 193 (2008) 10 - 22

FLOQUET THEORY

- NMR spectroscopy deals with time-dependent perturbations of nuclear spin systems and it is often imperative to solve the time-dependent Schrodinger Eq. in order to understand and predict evolution of these systems.

- In 1965, Shirley introduced Floquet theory to spectroscopy for solving the Schrodinger Eq. with a Hamiltonian representing periodically time-dependent interactions.

- Shirley replaced the Hilbert space finite-dimensional time-dependent Hamiltonian by an infinite-dimensional time-independent Floquet Hamiltonian and showed how it can be used to obtained an exact solution to the LvN equation.

- FT was extended to NMR by Vega and Maricq.
- It provides a more universal approach for the description of the full time dependence of the response of a periodically time-dependent system.
- It allows the computation of the full spinning sideband pattern that is of importance in many MAS experimental circumstances to obtain information on anisotropic sample.

- Shirley's Floquet formalism is also used in atomic and molecular spectroscopy methods.

The Floquet theory is exploited in many situations in NMR for example:

- *Time dependent periodic magnetic field*
- *Sample spinning*

FLOQUET THEORY

$$i\frac{dU}{dt} = H(t)U(t)$$

H (t) is a complex n by n matrix-valued function

Initial condition $U(0) = I$

Solution $U(t) = P(t)e^{-itF}$

where P(t) = P(t + T) and F is constant

The Structure of U(t) can be use in two independent theoretical approaches

1. **Fourier expansion** of the formal solution, leading to an infinite system of LDE with constant coefficient. The price of this approach is to handle an infinite dimension that can only be resolved numerically by truncation.

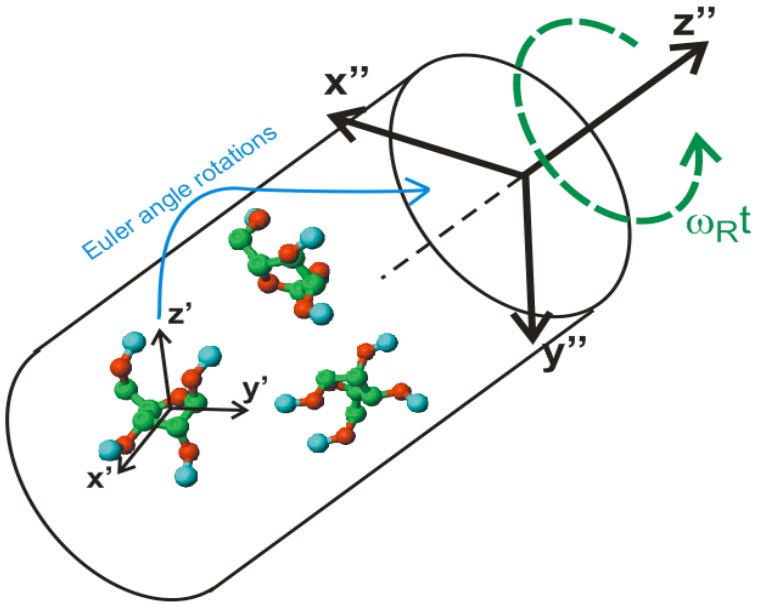
2. **Perturbation Nature**: $P(t) = \sum_{n=1}^{\infty} P_n(t)$ **[31]**

$$F = \sum_{n=1}^{\infty} F_n \quad \textbf{[32]}$$

Each term F_n is fixed such that $P_n(t) = P_n(t+T)$, which in turn ensures the Floquet structure at any order of approximation.

Applications of Floquet Theory

- **Variety of magnetic resonance phenomena:**
  - (1) Design of RF schemes of all kinds
  - (2) Calculation of the intensity of the centre bands and the sidebands in MAS spectra



Schematic depiction of an MAS rotor, showing rotor-fixed axes {x'',y'',z''} and randomly oriented molecules with molecule-fixed axes {x',y',z'}. The two axis systems are related by rotations by Euler angles, which are different for different molecules. The MAS rotor rotates about its z'' axis.

- **Multiphoton effects in NMR**
- **Electron paramagnetic resonance**
- **Nuclear quadrupole resonance**

Eugene S. Mananga, Application of NMR Spectroscopy, Bentham Science Publishers (2013)

C. Alternative Expansion Approaches Used Methods in Solid-State NMR

- Fer Expansion
- Floquet-Magnus Expansion

FER EXPANSION in NMR and Some its Applications

Fer expansion expresses the solution of Schrödinger equation in the form of an infinite-product of a series of exponentials

$$U(t) = \prod_{k=1}^{\infty} e^{F_k(t)} = e^{F_1(t)} e^{F_2(t)} \ldots .$$

For time-independent H or when H is such that $\left[\int_0^t dt' H(t), H(t)\right] = 0$, we get

$$F_1 = \int_0^t dt' H(t'),$$

such that the evolution operator $U(t) = e^{F_1(t)}$. Hence, $\frac{F_1(\tau_c)}{\tau_c} = \overline{H}$, in the sense of Magnus expansion. τ_c is the period of H.

$$U(t) = e^{F_1(t)} U_1(t); \qquad U_1(0) = I.$$

The above procedure may be iterated, and after n iterations we get the following:

$$U = e^{F_1} e^{F_2} \ldots e^{F_n} U_n, \qquad U_1(0) = I,$$

$$F_n = -i^n \int_0^t dt' H_F^{(n-1)}(t'), \qquad H_F^{(0)} = H,$$

$$H_F^{(n)} = e^{-F_n} H^{(n-1)} e^{F_n} - \int_0^1 dx e^{-xF_n} H e^{xF_n}.$$

due to Wilcox [112], a compact expression for $H_F^{(n)}$ may be obtained as

$$H_F^{(n)} = \sum_{k=1}^{\infty} \frac{(-1)^k k}{(k+1)!} \left\{ F_n^k, H_F^{(n-1)} \right\}$$

$$= -\frac{1}{2} \left[F_n, H_F^{(n-1)} \right] + \frac{1}{3} \left\{ F_n^2, H_F^{(n-1)} \right\} \ldots$$

where $n = 1, 2, 3, \ldots$. The second term on the right-hand side of Eq. (1

$$\left\{ F_n^2, H_F^{(n-1)} \right\} = \left[F_n, \left[F_n, H_F^{(n-1)} \right] \right].$$

- **The propagator for the FE with the 0th and 1st-order average Hamiltonians is given by**

$$U(\tau_C) = e^{-i\tau_C \overline{H_{Fer}^{(0)}}} e^{-i\tau_C \overline{H_{Fer}^{(1)}}}.$$

- **The first few terms in the nth-order Hamiltonian are defined as**

$$H_n(t) = H_{n,0}(t) + H_{n,1}(t) + H_{n,2}(t) + \cdots,$$

with

$$H_{n,0}(t) = -\frac{1}{2}[F_n(t), H_{n-1}(t)],$$

$$H_{n,1}(t) = \frac{1}{3}[F_n(t), [F_n(t), H_{n-1}(t)]],$$

$$H_{n,2}(t) = -\frac{1}{8}[F_n(t), [F_n(t), [F_n(t), H_{n-1}(t)]]],$$

$$\cdots$$

$$\cdots$$

$$\cdots$$

where $F_{n,j}(t)$ is defined as

$$F_{n,j}(t) = -i\int_0^t H_{n-1,j}(t')dt'$$

and the corresponding average Hamiltonian is defined as

$$\overline{H_{Fer}^{(n-1,j)}} = \frac{i}{\tau_C} F_{n,j}(\tau_C).$$

Heteronuclear dipolar decoupling

$$H(t) = d_{IS} S_Z (I_Z \cos \omega_{1I} t - I_Y \sin \omega_{1I} t) - \Delta\omega (I_Z \sin \omega_{1I} t - I_Y \cos \omega_{1I} t)$$

where d_{IS} is the dipolar coupling constant. $\Delta\omega$ is the off-resonance value, and ω_{1I} is the nutation frequency of the RF irradiation on the I spins. $H(t)$ is periodic with period $\tau_C = \frac{2\pi}{\omega_{1I}}$. A direct integration of Eq. (92) gives

$$\overline{H^{(0)}_{AHT/FT}} = \overline{H^{(0)}_{Fer}} = 0, \qquad \text{and} \qquad \overline{H^{(1)}_{AHT/FT}} = \overline{H^{(1,0)}_{Fer}}$$

$$\overline{H^{(2)}_{AHT/FT}} = \overline{H^{(1,1)}_{Fer}}.$$

$$U(\tau_C) \approx \exp\left\{-i\tau_C(\overline{H^{(1)}_{AHT/FT}} + \overline{H^{(2)}_{AHT/FT}} + \cdots)\right\}$$

$$\approx \exp\left\{-i\tau_C(\overline{H^{(1,0)}_{Fer}} + \overline{H^{(1,1)}_{Fer}} + \overline{H^{(1,2)}_{Fer}} + \cdots)\right\}.$$

$$\overline{H^{(i)}_{AHT/FT}} = \overline{H^{(1,i-1)}_{Fer}} \text{ with } i = 1, 2, \ldots, 5.$$

Cross polarization

$$H = dI_Z S_Z + \omega_1 I_X + \omega_1 S_X .$$

The applicable Hamiltonian in the interaction frame defined by the propagator

$$U_{RF}(t) = \exp\{-i\omega_1 (I_X + S_X)t\}$$

is written as

$$\tilde{H}(t) = \frac{d}{2}\{I_Z S_Z + I_Y S_Y + (I_Y S_Z + I_Z S_Y)\sin 2\omega_1 t + (I_Z S_Z - I_Y S_Y)\cos 2\omega_1 t\} .$$

The 0th, 1st, and 2nd-order average Hamiltonians for the Fer expansion are calculated to be

$$\overline{H^{(0)}_{Fer}} = \frac{d}{2}(I_Z S_Z + I_Y S_Y) = \overline{H^{(0)}_{AHT/FT}}$$

$$\overline{H^{(1,0)}_{Fer}} = \frac{d^2}{16\omega_1}(I_X + S_X) = \overline{H^{(1)}_{AHT/FT}}$$

$$\overline{H^{(1,1)}_{Fer}} = \frac{d^3}{64\omega_1^2}(I_Z S_Z - I_Y S_Y) = \overline{H^{(2)}_{AHT/FT}} .$$

The propagators for CP can be written as

$$U_{AHT/FT}(\tau_C) \approx \exp\left[-i\tau_C\left\{\frac{d}{2}(I_Z S_Z + I_Y S_Y) + \frac{d^2}{16\omega_1}(I_X + S_X) + \frac{d^3}{64\omega_1^2}(I_Z S_Z - I_Y S_Y) + \cdots\right\}\right]$$

$$U_{Right\text{-}run\text{-}FE}(\tau_C) \approx \exp\left\{-i\tau_C\frac{d}{2}(I_Z S_Z + I_Y S_Y)\right\}\exp\left[-i\tau_C\left\{\frac{d^2}{16\omega_1}(I_X + S_X) + \frac{d^3}{64\omega_1^2}(I_Z S_Z - I_Y S_Y)\right\}\right]\cdots$$

$$U_{Left\text{-}run\text{-}FE}(\tau_C) \approx \exp\left[-i\tau_C\left\{-\frac{d^2}{16\omega_1}(I_X + S_X) + \frac{d^3}{64\omega_1^2}(I_Z S_Z - I_Y S_Y)\right\}\right]\exp\left\{-i\tau_C\frac{d}{2}(I_Z S_Z + I_Y S_Y)\right\}\cdots$$

Bloch–Siegert shift

- **The untruncated rotating frame Hamiltonian during an on-resonance RF irradiation with**

$$H_{RF} = -2\omega_1 I_X \cos(\omega t + \phi)$$

is given by

$$H = H_R^{on}(t) = -\Delta\omega I_Z - \omega_1 [I_X \cos\phi + I_Y \sin\phi] - \omega_1 [I_X \cos(2\omega_I t - \phi) + I_Y \sin(2\omega_I t - \phi)]$$

$$F_1(t) = \int_0^t H dt = -\Delta\omega I_Z t - \omega_1 t [I_X \cos\phi - I_Y \sin\phi]$$

$$- \frac{\omega_1}{2\omega_I} \{I_X [\sin(2\omega_I t - \phi) + \sin\phi] + I_Y [\cos(2\omega_I t - \phi) - \cos\phi]\}$$

$$H_F^{(0)} = H$$

$$H_F^{(1)} = \sum_{k=1}^{\infty} \frac{(-1)^k k}{(k+1)!} \{F_1^k(t), H\}$$

- **The calculated coefficient term of (−IZ) is found to be the well documented Bloch–Siegert shift given by**

$$\omega_I \left(\frac{\omega_I}{2\omega_I}\right)^2$$

FLOQUET - MAGNUS EXPANSION in NMR and Quantum Physics

$$i\frac{dP}{dt} = H(t)P(t) - P(t)F$$

F is an unknown constant matrix determined with P (t+T) = P (t) to preserve the time periodicity

Using the expo. ansatz $P(t) = e^{-i\Lambda(t)}$

FME: $$\frac{d\Lambda}{dt} = \sum_{k=0}^{\infty} \frac{B_k}{k!}(-i)^k (ad_\Lambda)^k \left(H(t) + (-1)^{k+1} F\right)$$

Note: **ME** and **FME** equ. are similar but with the addition of **F**

ME: $$\frac{d\Omega}{dt} = \sum_k \frac{B_k}{k!}(-i)^k ad_\Omega^k (H(t))$$

Eugene Mananga and Thibault Charpentier, JCP, 135 (2011) 044109

The FME equations are independent of $\Lambda(0)$

The advantage of the FME approach is its ability to make choices for $\Lambda(0)$ different from the generally assumed $\Lambda(0)=0$ simplifying the perturbative calculation of $\Lambda(t)$ and F .

The choice of: $\Lambda(0)\neq 0$

is equivalent to the use of a more general representation of the evolution operator

Floquet-Magnus Operator: $U(t)=P(t)e^{-itF}P^{+}(0)$

This removes the constraint of a stroboscopic observation

Floquet Operator: $U(t)=P(t)e^{-itF}$

Eugene Mananga and Thibault Charpentier, JCP, 135 (2011) 044109

The link between the Floquet-Magnus and Magnus Expansions can be obtained from:

$$U(T,0)=\exp(-i\Omega(T))=\exp\left(-iT\,e^{-i\Lambda(0)}\,F\,e^{i\Lambda(0)}\right)$$

where $$\frac{\Omega(T)}{T}=e^{-i\Lambda(0)}\,F\,e^{i\Lambda(0)}$$

Only in the case $\Lambda(0)=0$ that **FME** provides **AHT (ME)** results:

$$\frac{\Omega(T)}{T}=F$$

However, **ME** is limited to the construction of the **AHT**, whereas the **FME** also constructs the operator $\Lambda(t)$ providing the means to obtain the evolution of the system in between the stroboscopic detection points.

Eugene Mananga and Thibault Charpentier, JCP, 135 (2011) 044109

The General Formula Developed For The FME Are:

$$\Lambda_n(t) = \int_0^t G_n(\tau)\, d\tau - tF_n + \Lambda_n(0)$$

$$F_n = \frac{1}{T} \int_0^T G_n(\tau)\, d\tau$$

Functions are constructed using the FME recursive generation scheme.

If higher order terms can be easily computed numerically, we believe that symbolic calculations software can enable formal derivation of higher order terms.

Eugene Mananga and Thibault Charpentier, JCP, 135 (2011) 044109

1. Application of the FME on a Simple Hamiltonian
- Static perturbation theory versus FME -

Here, we compare the FME to the SPT which has been shown to yield the correct form of Zeeman truncated NMR interactions without the limit of stroboscopic observation of the AHT. This will give us the opportunity to shed a new light on the FME scheme and the derivation of a criterion for the two theories being compatible.

CONSIDER THE COMMON FORM OF HAMILTONIAN IN SS-NMR

$$H = \omega_0 I_Z + \lambda \sum_m (-1)^m R_{2,-m} T_{2,+m}$$

FME provides an expansion which is in agreement with the SPT and the VVT. This is not the case of the ME

SPT

- $\omega_0 I_Z$ is the Zeeman interaction
- $R_{2,m}$ are the lattice parts of the internal interaction
- $T_{2,m}$ are second rank m-order spherical tensor describing the spin system

$$H_{SPT} = \omega_0 I_Z + \lambda R_{2,0} T_{2,0} + \frac{\lambda^2}{2\omega_0} \sum_{m \neq 0} \frac{R_{2,m} R_{2,-m}}{m} \left[T_{2,m}, T_{2,-m} \right]$$

Eugene Mananga and Thibault Charpentier, JCP, 135 (2011) 044109

2. Extension of FME to Multimode Hamiltonian

STRAIGHTFORWARD:

Considering the generalized Fourier expansion of the Hamiltonian:

$$H(t)=\sum_{\vec{m}} H_{\vec{m}} \exp\left(-i\,\vec{m}\cdot\vec{\omega}\,t\right)$$

First Order terms:

$$\Lambda_1(t)=\sum_{\vec{m}\cdot\vec{\omega}\neq 0} \frac{H_{\vec{m}}}{i\,\vec{m}\cdot\vec{\omega}} e^{-i\,\vec{m}\cdot\vec{\omega}\,t}$$

$$F_1=\sum_{\vec{m}\cdot\vec{\omega}=0} H_{\vec{m}}$$

Calculation of second order terms is also straightforward using FME eqs. These expressions highlight the fact that the multimode Hamiltonian case can be treated in Hilbert space

Eugene Mananga and Thibault Charpentier, JCP, 135 (2011) 044109

3. Application of FME to Recoupling Sequences

The dipolar spin interaction MAS is given by

$$H_D(t) = \frac{1}{2}\sum_{i\neq j}\omega_D^{ij}(t)T_{20}^{ij},$$

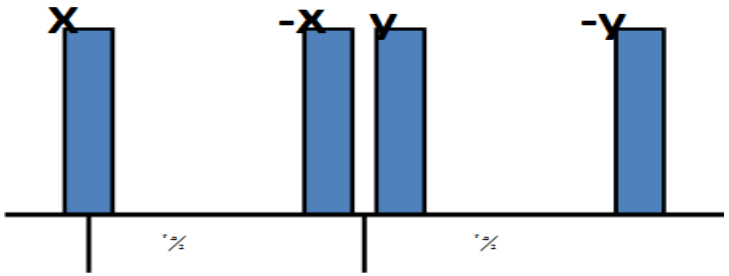
$$T_{20}^{ij} = \tfrac{1}{\sqrt{6}}\left[2I_{ZZ}^{ij} - I_{XX}^{ij} - I_{YY}^{ij}\right]$$

$$\omega_D^{ij}(t) = b_{ij}\sum_{n=-2}^{2} C_n^{ij}(\alpha^{ij},\beta^{ij},\gamma^{ij})e^{-in\omega_R t} = b_{ij}\sum_{n=-2}^{2} C_n^{ij}(\alpha^{ij},\beta^{ij})e^{-in(\omega_R t+\gamma^{ij})}$$

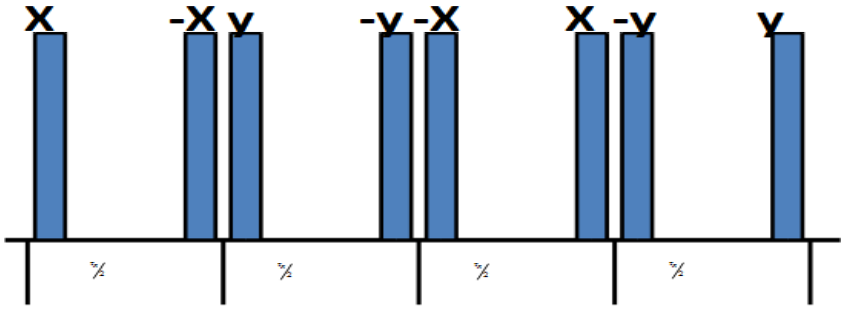
$$C_n^{ij}(\alpha^{ij},\beta^{ij}) = d_{0,n}^2(\theta_M)\sum_{n'=-2}^{2}(-1)^{n'}Y_{2n}^{ij},e^{-in\alpha^{ij}}d_{n-n'}^2(\beta^{ij})$$

$$\varphi = t/\tau_R$$

BABA I

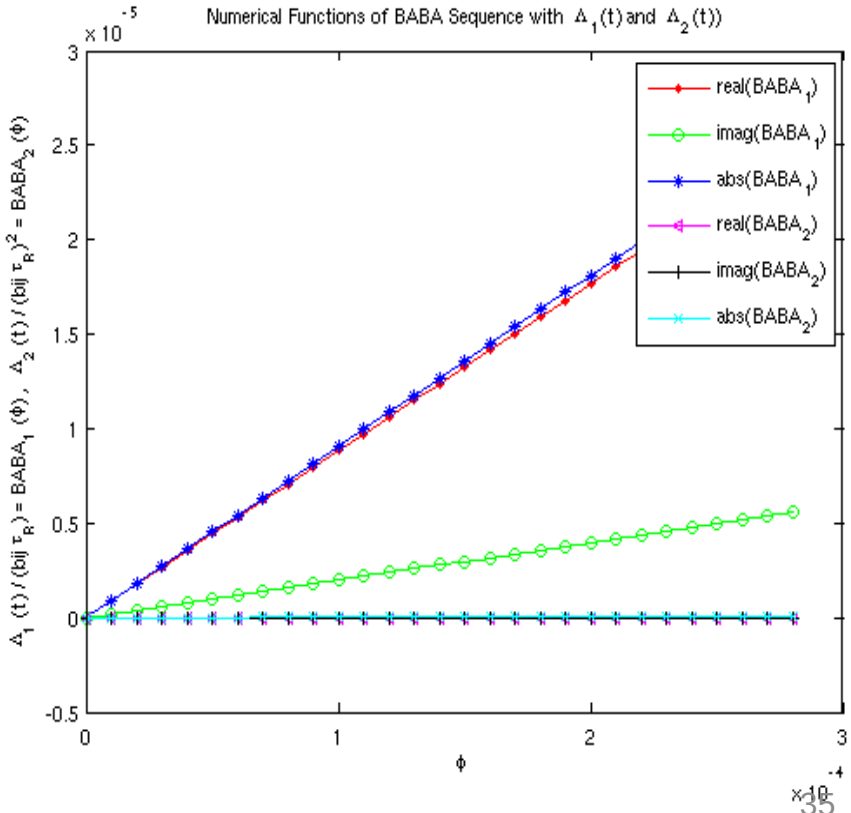


BABA II

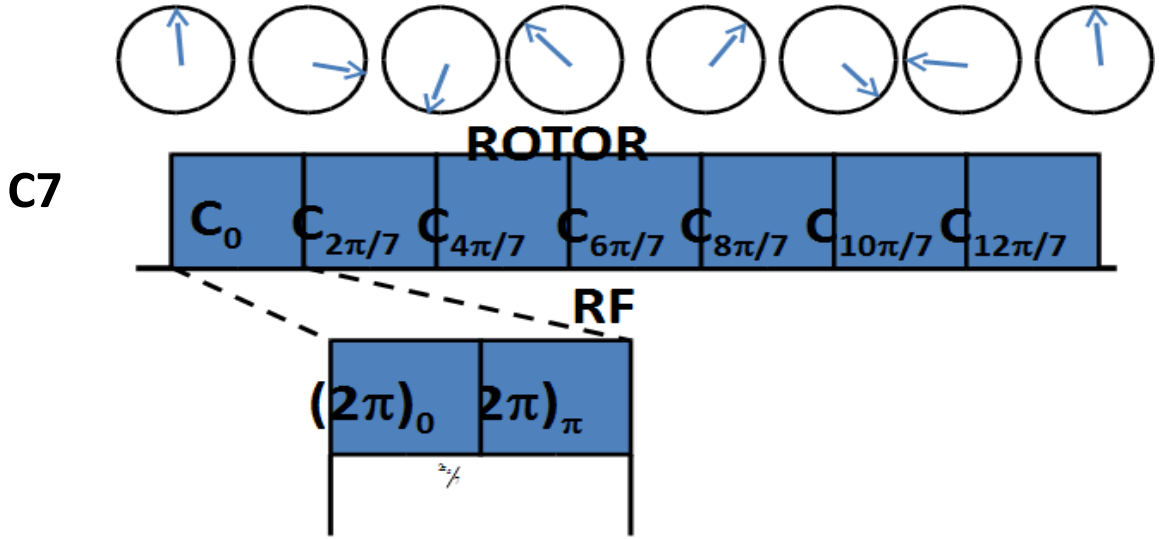


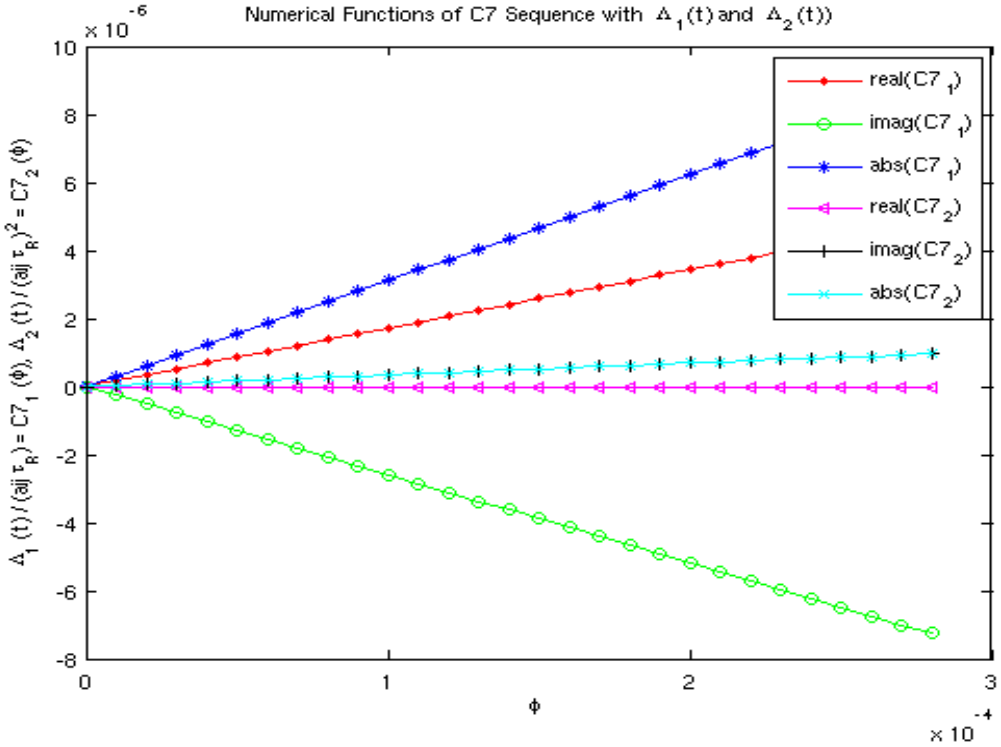
$$\frac{\Lambda_1(t)}{b_{ij}\tau_R} = BABA_1(\varphi) = \frac{1}{16\sqrt{2}\pi}\left(i - \frac{1}{\pi\sqrt{2}}\right)(e^{-i2\pi\varphi}-1)(I_{YY}^{ij}-I_{XX}^{ij})$$

$$\frac{\Lambda_2(t)}{b_{ij}^2\tau_R^2} = BABA_2(\varphi) = \left\{\frac{1}{768}\left[\frac{-1}{2}(e^{-4\pi i\varphi}-1)+(e^{-i2\pi\varphi}-1)\right]\right.$$
$$\left.+\frac{i}{768\pi^3}\left[\frac{1}{8\sqrt{2}}(e^{-i4\pi\varphi}-1)-(e^{-i2\pi\varphi}-1)\right]\right\}(I_{YY}^{ij}-I_{XX}^{ij})$$
$$-\left\{\frac{1}{6144\pi^2}(1-\frac{1}{2\sqrt{2}})(e^{i2\pi\varphi}+e^{-i2\pi\varphi}-2)\right\}(I_{YY}^{ij}-I_{XX}^{ij}),$$



Eugene Mananga and al., SSNMR, 41, 32-47 (2012)



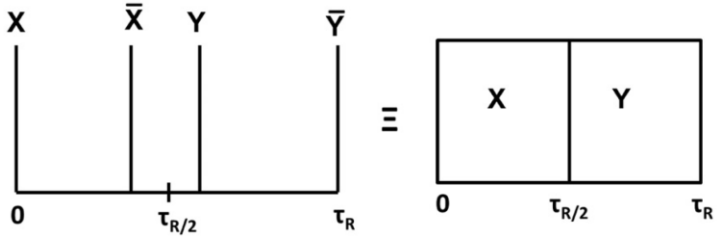


$$\|\overline{H}^{0}_{DQ}\|_{BABA} \simeq 1.4 \sin\gamma^{ij} \|\overline{H}^{0}_{DQ}\|_{C7}$$

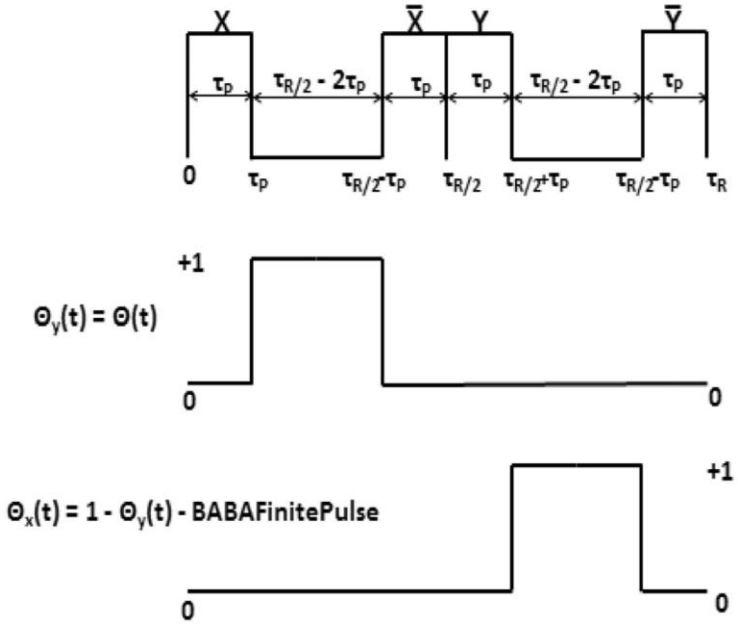
Eugene Mananga and al., SSNMR, 41, 32-47 (2012)

4. Application of FME to Finite Pulse Sequences

BABA pulse sequence with δ-pulse width.



BABA pulse sequence with finite pulse width



Two important findings of the above result should be recognized.

- **First, when the pulse width = 0, F_1 is reduced to the build-up DQ coherence expression:**

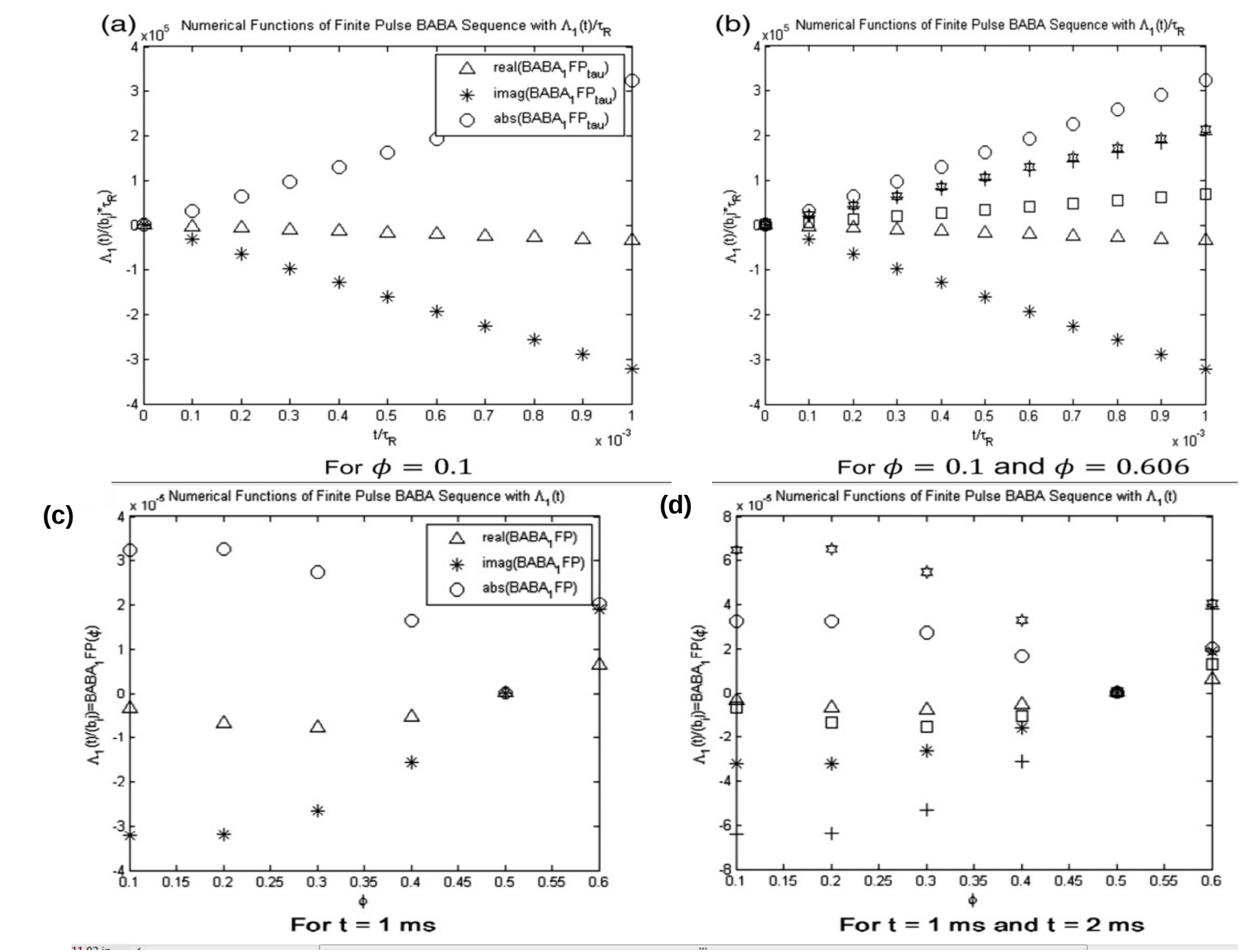
$$F_1 = \bar{H}^0 = \frac{3}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left(\frac{1}{2}\right) \left(I_{YY}^{ij} - I_{XX}^{ij}\right)$$

- **Second, The expression of F_1 that describes not only the build-up but also the destruction Of DQ coherence can also be obtained:**

$$F_1 = \bar{H}^0 = \frac{3}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left(\frac{1}{2} - \emptyset\right) (I_{YY}^{ij} - I_{XX}^{ij})$$

$$\emptyset = \frac{2\tau_P}{\tau_R}$$

Eugene Mananga and A. Reid, Mol. Phys., 111, 2, 243-257, (2013)



Eugene Mananga and A. Reid, Mol. Phys., 111, 2, 243-257, (2013)

APPLICATIONS OF FLOQUET-MAGNUS EXPANSION IN PHYSICS

Rigorous Bound on Energy Absorption and Generic Relaxation in Periodically Driven Quantum Systems

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We discuss the universal nature of relaxation in isolated many-body quantum systems subjected to global and strong periodic driving. Our rigorous Floquet analysis shows that the energy of the system remains almost constant up to an exponentially long time in frequency for arbitrary initial states and that an effective Hamiltonian obtained by a truncation of the Floquet-Magnus expansion is a quasi-conserved quantity in a long timescale. These two general properties lead to intriguing classification on the initial stage of relaxation, one of which is similar to the prethermalization phenomenon in nearly-integrable systems.

PHYSICAL REVIEW A **91**, 020101(R) (2015)

Floquet resonant states and validity of the Floquet-Magnus expansion in the periodically driven Friedrichs models

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The Floquet eigenvalue problem is analyzed for periodically driven Friedrichs models on discrete and continuous space. In the high-frequency regime, there exists a Floquet bound state consistent with the Floquet-Magnus expansion in the discrete Friedrichs model, while it is not the case in the continuous model. In the latter case, however, the bound state predicted by the Floquet-Magnus expansion appears as a metastable state whose lifetime diverges in the limit of large frequencies. We obtain the lifetime by evaluating the imaginary part of the quasienergy of the Floquet resonant state. In the low-frequency regime, there is no Floquet bound state and instead the Floquet resonant state with exponentially small imaginary part of the quasienergy appears, which is understood as the quantum tunneling in the energy space.

PHYSICAL REVIEW X **4,** 031027 (2014)

Periodically Driven Quantum Systems: Effective Hamiltonians and Engineered Gauge Fields

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Driving a quantum system periodically in time can profoundly alter its long-time dynamics and trigger topological order. Such schemes are particularly promising for generating nontrivial energy bands and gauge structures in quantum-matter systems. Here, we develop a general formalism that captures the essential features ruling the dynamics: the effective Hamiltonian, but also the effects related to the initial phase of the modulation and the micromotion. This framework allows for the identification of driving schemes, based on general N-step modulations, which lead to configurations relevant for quantum simulation. In particular, we explore methods to generate synthetic spin-orbit couplings and magnetic fields in cold-atom setups.

Floquet-Magnus Theory and Generic Transient Dynamics in Periodically Driven Many-Body Quantum Systems

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This work explores a fundamental dynamical structure for a wide range of many-body quantum systems under periodic driving. Generically, in the thermodynamic limit, such systems are known to heat up to infinite temperature states after infinite-time evolution, irrespective of dynamical details. In the present study, instead of considering infinitely long-time scale, we aim to provide a framework to understand the *long but finite time* behavior, namely the transient dynamics. In the analysis, we focus on the Floquet-Magnus (FM) expansion that gives a formal expression of the effective Hamiltonian on the system. Although in general the full series expansion is not convergent in the thermodynamics limit, we give a clear relationship between the FM expansion and the transient dynamics. More precisely, we rigorously show that a truncated version of the FM expansion accurately describes the exact dynamics for a finite-time scale. Our result reveals a reliable time scale of the validity of the FM expansion, which can be comparable to the experimental time scale. Furthermore, we discuss several dynamical phenomena, such as the effect of small integrability breaking, efficient numerical simulation of periodically driven systems, dynamical localization and thermalization. Especially on thermalization, we discuss generic scenario of the prethermalization phenomenon in periodically driven systems.

Brillouin-Wigner theory for high-frequency expansion in periodically driven systems: Application to Floquet topological insulators

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We construct a systematic high-frequency expansion for periodically driven quantum systems based on the Brillouin-Wigner (BW) perturbation theory, which generates an effective Hamiltonian on the projected zero-photon subspace in the Floquet theory, reproducing the quasienergies and eigenstates of the original Floquet Hamiltonian up to desired order in $1/\omega$ with ω being the frequency of the drive. The advantage of the BW method is that it is not only efficient to derive higher order terms, but it is even possible to formally write down the whole infinite series expansion, as compared to the van Vleck degenerate perturbation theory. The expansion is also free from a spurious dependence on the driving phase, which has been an obstacle in the Floquet-Magnus expansion. We apply the BW expansion to various models of noninteracting electrons driven by circularly polarized light. As the amplitude of the light is increased, the system undergoes a series of Floquet topological-to-topological phase transitions, whose phase boundary is well explained by the BW expansion in the high-frequency regime. As the frequency is lowered, the high-frequency expansion breaks down at some point due to band touching with non-zero photon sectors, where we find, numerically, even more intricate and richer Floquet topological phases spring out. We have also analyzed, with the Floquet dynamical mean-field theory, how the effects of electron-electron interaction and energy dissipation appear. Specifically, we find phase transitions from Floquet-topological to Mott insulators emerge, where the phase boundaries can again be understood by the high-frequency expansion.

Comparison between Fer and Floquet-Magnus expansions in Solid-State NMR and Physics

$$H(t) = \sum_{m \neq 0} H_m e^{-im\omega t} + H_0$$

$$F_{FME}^{(0)} = \overline{H_{FE}^{(0)}} = H_0$$

$$F_{FME}^{(1)} = \overline{H_{FE}^{(1,0)}} = \sum_{m \neq 0} \frac{[H_{-m}, H_m]}{2m\omega} + \sum_{m \neq 0} \frac{[H_m, H_0]}{m\omega}$$

The discrepancy between the FME and FE approaches arises at higher orders. For instance, we see that the second order expansions of both schemes are different. The second order term of the FME (Eq. (163)) has more terms and it looks much more complicated than the 2nd-order term of the Fer expansion. From the computational effectiveness point of view, this tells us that the rate of convergence of the FE is faster than that of the FME, for a prescribed precision, one needs more Λ_n's (FME) than F_k's (FE). From the computational point of view, in the sense of spin dynamics with fast-oscillation Hamiltonian, the FME requires more work than the FE. Hence, the characteristics of the problem at hand might eventually dictate the method to be used.

CONCLUSION

Schrodinger picture LVN: $$i\frac{dU}{dt} = H(t)U(t)$$

AHT: $$U(t_c) = \exp\left\{-i\overline{H}(t_c)t_c\right\}$$

FLT: $$U(t) = P(t).\exp\left\{-iH_F t\right\}$$

Connection FLT and AHT: $$P(0) = P(nt_c) = 1 \qquad H_F = \overline{H}$$

FME: $$U(t) = P(t)\exp\left\{-itH_F\right\}P^{+}(0)$$

Connection FME and FLT: $$P(t) = \exp\left\{-i\Lambda(t)\right\}$$

FE: $$U(t) = \prod_{k=1}^{\infty}\exp\left\{H_{F_k}(t)\right\} = \exp\left\{H_{F_1}(t)\right\}\exp\left\{H_{F_2}(t)\right\}...$$

SPT: $$U(t) = \sum_{m} U_m(t)\exp(im\omega t)$$

We mainly presented the **FLOQUET-MAGNUS EXPANSION** useful to shed new lights on **AHT** and **FT.**

The theory is based on two operators:

$\Lambda(t)$ Describes the evolution within the period

F The Hamiltonian governing the evolution at multiple of the period

A crucial parameter has been shown to be periodic boundary condition $\Lambda(0)$

FME Theory can be directly connected to the **AHT** for $\Lambda(0)=0$ yielding $F=H_{AHT}$

But, in constrast to the ME, the knowledge of $\Lambda(t)$ allow the evolution in-between the stroboscopic points to be evaluated

- Equivalence with FT is obtained from a special choice of $\Lambda(0) \neq 0$ leading to an expansion that is equivalent with the SPT.

- **FME** provides a means to calculate higher order terms allowing the disentanglement of the stroboscopic observation $\Lambda(t)$ and effective Hamiltonian F that will be useful to describe spin dynamics in SS-NMR.

- **FME** offers a simple way to handle multiple incommensurate frequencies and thus open the perspectives to deal with multi-mode Hamiltonian in the Hilbert space.

FME in SSNMR can provide new aspects not present in AHT and FT such as recursive expansion scheme in Hilbert space that can facilitate the devise or improvement of pulse sequences

It is expected that the FME will provide means for more accurate and efficient Spin dynamics simulation and for devising new RF pulse sequence

The Possibility of Enhanced FME Performance Certainly Deserves Further Attention and Additional Quantitative Work Would Demonstrate the Utility of the Approach in Nuclear Magnetic Resonance Spectroscopy

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Thank you
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